RADIOACTIVITY IN THE MARINE ENVIRONMENT

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^{54}Mn, and ^{55,59}Fe. The MIKE* thermonuclear explosion in 1952 generated considerable transuranium isotopes, including higher mass isotopes of plutonium. A sensible fraction of the debris from MIKE was deposited in the Pacific, thus providing a number of unique tracers with fairly localized input, such as ^{241}Pu and ^{242}Pu.

It appears that approximately 10^{28} atoms of ^{239}Pu (and perhaps one tenth as much ^{240}Pu) have been generated by thermonuclear explosions in the course of testing. Neptunium-237 has also been produced in quantities comparable to ^{239}Pu.

One hundred and forty megatons, or 72 percent of the total yield, were produced as airbursts, i.e., under conditions in which the fireball did not intersect the ground. Documented cases of tower-burst inputs represent a small fraction of the total—in the vicinity of 0.5 percent—and may be taken to be of trivial significance in the context of the marine environment. However, the third category, surface bursts, while representing only about 28 percent of the total, deserves special attention because of highly localized input, the majority of which was in the Marshall Islands, in the 1952 Ivy, 1954 Castle, 1956 Redwing, and 1958 Hardtack series.

Characteristics of Radioactive Debris

The characteristics of the particles formed in a nuclear explosion are determined by a large number of factors. Among the more important are the yield of the device and the quantity and kind of nearby environmental materials interacting with the nuclear explosion. The biological availability of the radioactivity in the debris particles will depend on particle size, the chemical form of the radioelement, and the matrix in which the radioactivity is imbedded or to which it is attached.

PARTICLE SIZE DISTRIBUTION

Airbursts In general, more than 90 percent of the radioactivity in airburst debris is to be found in very finely divided particles, less than 1 μ in diameter. The literature on the particle size distribution of debris produced by airbursts is sparse. Nathans suggests a log-normal size distribution with a peak diameter in the differential size distribution below 1 μ in diameter (M. Nathans, Tracerlab Inc., personal communication). Sherer proposed an exponential form of distribution for particles larger than 1 μ, based on microscopic sizing of particles of verified radioactivity, e.g., the particle size distribution as a function of its diameter is proportional to $e^{-D/b}$, where b is a parameter having a value of 1-2 μ^{-1} for nominal-size (20-kt) explosions and D is the diameter in microns (J. Sherer, Lawrence Radiation Laboratory, Livermore, Calif., personal communication).

Whatever the true form of the size distribution function, it is clear that the particle population increases rapidly with decreasing particle size to below 1 μ. There is considerable indirect evidence that particle size is related to the yield of the device, or more precisely, to the yield to mass ratio. The size distribution curve is shifted to smaller sizes as the yield increases. Since the bulk of the radioactivity in airbursts resides in submicron particles, and these are slowly acted upon by gravitational forces, the worldwide stratospheric distribution of airburst debris is relatively insensitive to the exact form of the size distribution. Very few particles in excess of 10 μ in diameter are found at any yield.

Tower Bursts In tower bursts, a considerable portion of the supporting structure will be volatilized or melted. Few quantitative data have been published that reveal details of the size distribution of tower burst events. It is clear from environmental studies accompanying tests, however, that the particle population contains particles much larger than are found in airbursts of the same yield. For example, Larson (1966) reports that significant fractions of the radioactivity are to be found in particles in excess of 44 μ. Hence, a large fraction of the radioactivity is deposited locally, and proportionately less enters the long-range-long-term atmospheric distribution system.

Coral Island Surface Bursts A number of explosions of moderately high to high yield have been conducted on coral island surfaces or on barges, sometimes in relatively shallow water. In these cases substantial quantities of coral may be melted and vaporized, and, together with the vaporized constituents of seawater, may become major radioactive debris constituents.

Data on the size distribution of radioactive debris from multimegaton explosions on coral islands are limited. The sheer magnitude of the event precludes collection of adequate samples from the prompt fallout and the residual nuclear cloud. The samples that have been retrieved and analyzed inevitably suffer from lack of definition concerning the precise fraction of the total radioactive particle population the sample represents.

Figure 1 gives a composite size distribution of radioactive particles for BRAVO as obtained from prompt fallout recorded on nearby islands. Note that the maximum observed in the vicinity of 150 μ reflects only that particles smaller than 150 μ were incompletely deposited at the collection point because of their finite gravitational settling rates.

Nathans *et al.* (1970), in studying the size distribution of cloud samples of nuclear debris of BRAVO and other coral island surface explosions, found that the results for all particles (radioactive and inert) over the range 1-100 μ fit a power law of the form $f(d) \propto CD^{-n}$, where n may have the value of about 4. Power law distributions are rather com-

*Operational code names; a chronology is given in Glasstone (1964).

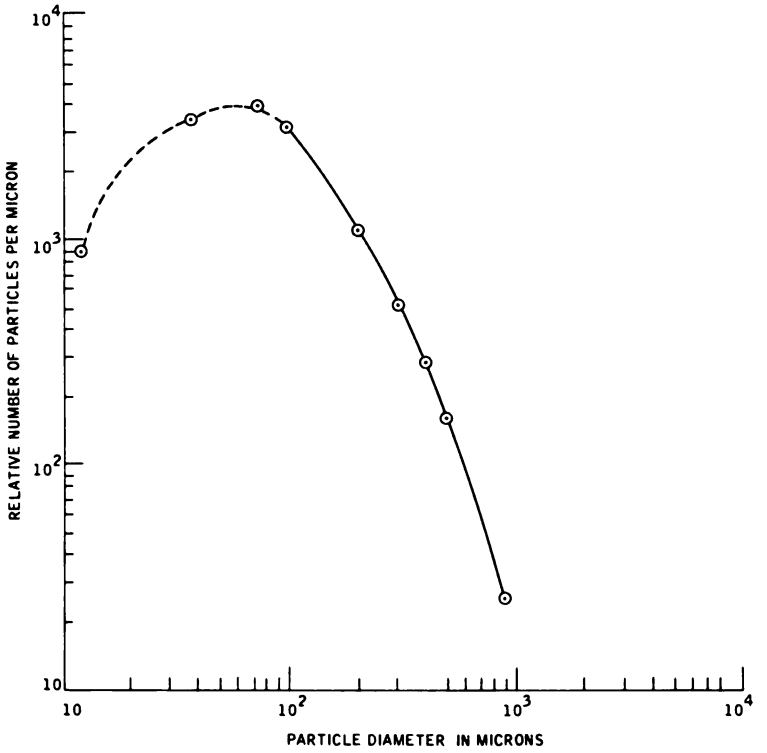

FIGURE 1 Composite differential particle size spectrum—BRAVO.

monly observed for natural aerosols, for zodiacal dust, and for stratospheric dust. Russell (1964) found a power law description for cloud samples of a low-yield surface explosion. Heft (1970) described a cloud from a coral surface explosion as representing the superposition of two log-normal distributions. However, because of the late time of sampling, Heft's samples, similar to Nathans', contained very little of the larger end of the particle size spectrum. In any event, there appears to be a physical basis for a multimode set of particle distribution functions describing the debris as a whole. At the larger end of the particle size spectrum, particles are formed by coalescence of smaller particles characteristic of the pre-shot soil. Melting and shock compaction are presumably partly responsible. A second phase, distributed about a smaller median diameter, is attributed to unmelted, still-crystalline debris swept into the nuclear clouds by the strong afterwinds.

ACTIVITY CONCENTRATIONS OF RADIOACTIVE DEBRIS

Radioactivity per unit mass of particle varies in both kind and amount with the size and shape of the particles of debris formed in nuclear tests. A term used to describe the radioactive content of particles is "activity concentration."* Activity of radioactive debris may be reported as atoms of radioactive species per gram of debris, as disintegrations per minute of radioisotope per gram, and so on. In this section, activity concentration is given in units of equivalent fissions per gram of debris. An equivalent fission is the number of fissile atoms (e.g., ^{235}U, ^{239}Pu, or ^{238}U) required to give the observed number of fission product atoms, or, alternatively, the number of fissioning atoms associated with a given number of product atoms of any kind.

The situation is complicated by a phenomenon called "fractionation." If the debris sample were unfractionated, i.e., representative of the true proportions in which the radioactivity was produced in the device, then equivalent fissions calculated for all isotopes in the sample will be identical. Because of differing chemical and physical properties, the elements may fractionate with respect to each other during the condensation and subsequent cooling phase in the fireball and nuclear cloud, or may fractionate by selective attachment onto pre-existing condensed materials. The elements in the chains that are gaseous at ordinary temperatures, or low-boiling, may experience sharp differentiations from elements whose metals or oxides are high-boiling. The more refractory elements or oxides (those condensing above 1500° K) will have condensed out well before 60 sec have elapsed, even for megaton explosions.

One can calculate the theoretical population of chain members in the first few minutes by using the charge division hypothesis of Glendenin *et al.* (1951), assigning probable nuclear charges based on the method of Wahl (1958), and using estimated half-lives after the method of Bolles and Ballou (1956).

*In view of a different use and connotation of "specific activity" in subsequent chapters, in this chapter we will use "activity concentration."

The average activity concentration of airburst debris is calculable if the fission yield and mass of the device structure are known. Denoting activity concentration as S,

$$\bar{S} = 1.45 \times 10^{17} (W/M).$$

Here W is the yield in kilotons and M is the mass in tons. The units of $\bar{S}$ are equivalent fissions per gram.

However, because of fractionation of the radioelements during the cooling phase, no specimen of debris may correspond to this value, except by chance. Benson *et al.* (1965) have presented data that imply that the activity concentration of airburst particles over the range of a few to 15 microns in diameter is roughly constant. They found that the ^{95}Zr content of the particles is roughly related to the cube of the particle diameter. Barium-140 shows a radial dependence, roughly as D^2: 3-2.6 over this size range. These larger particles are strongly depleted in ^{140}Ba and all other volatile or semivolatile chains. Hence, the bulk of their activity is contributed by zirconium and rare-earth nuclides and a few additional refractory or semirefractory species, leading to a quasiconstant activity concentration behavior.

Typically, it is found that the fraction of the mass found in particles in excess of 2 μ will exceed by a factor of three or more the fraction of the most refractory fission product found in this size interval. Thus, in the above example, 30 percent of the mass might be found in particles larger than 2 μ associated with only 10 percent of the ^{95}Ar. This result may be attributable to poor mixing between the volatilized device materials and the fission products. It may also arise from concentration effects, insofar as they modify the condensation temperature of the higher-boiling elements or their oxides. The fission products may represent only 10^{-3} to 10^{-4} of the concentration of the structural components in the fireball vapors.

In particles smaller than 2-3 μ, however, the activity concentration increases sharply for all nuclides, as required by mass and activity balance constraints. There are few published details on this region, and the general behavior must be inferred from gross beta particle specific activities.

In the case of surface or near-surface bursts, the expanding fireball will be admixed with inert soil, some of which will have been vaporized, some melted, and some of which will remain essentially unaltered. The extent and time sequence of mixing between inert debris and fission products will be variable from event to event and would be expected to be dependent upon explosion conditions. Soils are variable in composition, and water vapor can represent a significant and undetermined component of the system, particularly in coral surface bursts.

Despite the complexity of the chemical and physical processes attending a land-surface nuclear explosion, certain consistent patterns of debris behavior are observed.

Empirical observations of land-surface nuclear debris suggest that a distinction be made among at least three classes of particles.

1. Completely volatilized material that recondenses to finely divided particles consisting of the device structural members, the fission products, and some environmental material.
2. Melted but not completely volatilized environmental material into which fission products may diffuse or onto which they attach. The melted entities may coalesce to form larger particles.
3. Unmelted environmental material, swept into the nuclear cloud by the afterwinds, to which fission products or small condensed radioactive agglomerates attach by surface impaction.

To these may be added a fourth class–spherical particles of very high activity concentration (up to 10^{16} equivalent fissions per gram) are found. These may represent coalescence of condensed vaporized soil and fission product droplets, with rapid agglomeration to larger particles. Some coalescence of this component with melted inert droplets may also occur, leading to a spherical end product.

Figure 2 depicts empirical activity concentration versus particle size data for a 15-megaton coral island explosion. This curve is drawn on the basis of radioactive particles only. It was determined by radioautography that the fraction of radioactive particles diminished rapidly with decreasing size below 44 μ. This effect is responsible for the steep slopes of both classes of chains, refractory and volatile, below 44 μ. Consequently, the absolute magnitude of the slopes of the curves below 44 μ is not to be taken too literally, since there is some uncertainty in the assessment of the number of radioactive particles. If correction is not made for radioactive particles, the activity concentration curve for chain 89 is flat over the entire range of particle sizes.

In Table 4 are assembled some activity concentration data, expressed in equivalent fissions per gram of fallout, collected near the fallout site. Data for a refractory-chain ^{99}Mo, a volatile-chain ^{89}Sr, and a chain of intermediate volatility ^{140}Ba are given. These data represent activity concentrations that would be observed if the total yield of the device were due to fission, i.e., they have been normalized to 100 percent fission yield for comparative purposes. No attempt has been made to distinguish between radioactive and nonradioactive particles in the samples, with the exception of the 15-Mt case.

An admixture of inert (nonradioactive) debris will in general represent material physically displaced by the energy of the explosion to the sampling position.

Some significant points become apparent from a study of Table 4:

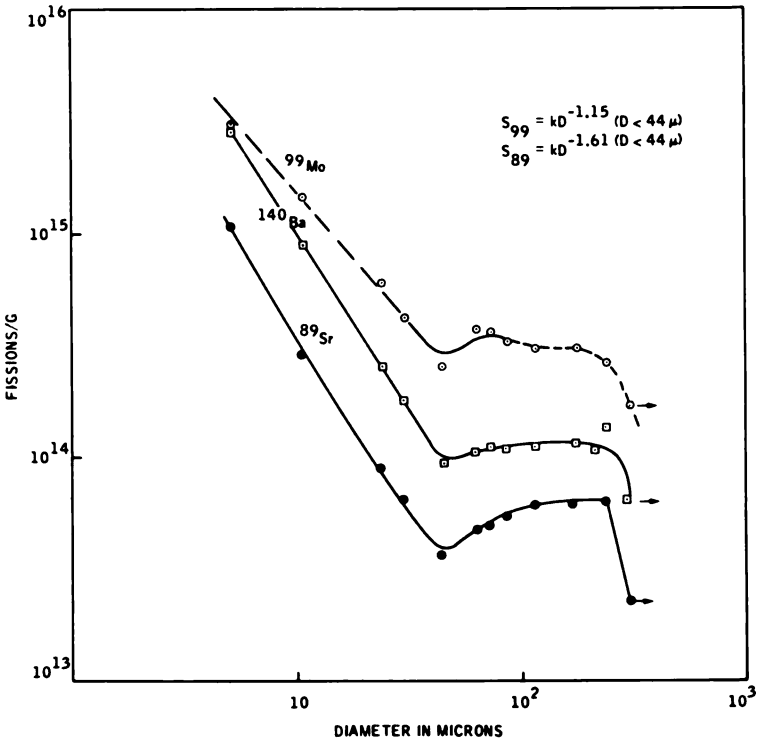

FIGURE 2 Empirical activity concentration *versus* particle size data for 15-megaton coral island explosion. S_{99}, activity concentration of chain 99; S_{89}, activity concentration of chain 89.

1. The normalized refractory-chain activity concentrations from coral surface explosions are relatively insensitive to yield, and, indeed, to particle size between about 50 μ and 250 μ in diameter. The average ^{99}F/g for the four events is 6.9×10^{14} over this range of sizes. The average deviation from the average is about 36 percent. The greatest variability is found at the large-particle end of the spectrum.

2. If columns 2 and 3 are compared, corresponding to fallout near zero and 80 km downwind, the activity concentration of the debris transported downwind is seen to be uniformly higher for all classes of isotopes. This suggests a longer opportunity for debris attachment processes to occur when the mean residence time in the cloud is lengthened. (However, absolute fallout intensities downwind will normally be lower because the number of grams of fallout deposited per unit area is smaller.)

3. Spherical particles (column 5) are uniformly higher in activity concentration for chain 99, a refractory chain, than are the irregular particles. [Tompkins and Krey (1956) report that about 25 percent of the total activity at 10 days after the explosion was attributable to spherical particles in the 5.1-Mt explosion, but only a few percent of the mass. Spherical particles were difficult to find in size fractions below 177 μ. One should not, however, infer their absence.]

4. Volatile chains show a somewhat greater variability in activity concentration from event to event than do refractory chains. The mean residence time in the cloud exerts a strong influence. (Compare 3.5 Mt near ground zero and 80 km downwind.) Also, the degree of mixing in the cloud appears to have been somewhat more effective in the 5.1-Mt case than in the other instances. Whereas spherical particles were strongly enriched in refractory chains relative to irregulars of the same size, this is not the case with volatile chains.

5. An activity concentration maximum in the vicinity of 300 μ is a recurring freature, which is most pronounced for the spherical population. Volatile chains appear to decrease in activity concentration roughly according to D^{-2} past 300 μ; the behavior is more in accord with D^{-1} for refractory chains, with the slope being slightly flatter for spheres than for irregulars. These features, if present in the 15-Mt case, were not revealed by the analysis that grouped all particles larger than 300 μ in a single fraction.

In summary, few unequivocal statements can be made concerning the activity concentration behavior of coral surface debris. However, certain of the observations have at least a partial explanation in the thermal and mixing history to which the debris was subjected.

We believe the large, high-specific-activity spheres to represent, in part, fireball condensate that coalesced to large droplets and, in part, melted soil particles that experienced no condensation history but that coalesced with high-activity-concentration droplets of condensate origin. Electron microprobe analysis of spheres from coral island explosions

TABLE 4 Activity Concentration and Particle Size, Coral Island Surface Explosions (Equivalent fissions $\times 10^{-14}$ per gram)[a]

$D_g(\mu)$	3.5 Mt (Prompt Fallout near Ground Zero)	3.5 Mt (Fallout Collected 80 km Downwind)	5.1 Mt	5.1 Mt (Spheres Only, near Ground Zero)	15 Mt (Radioactive Particles Only, Shot Atoll, Spheres plus Irregulars)	0.04 Mt (All Particles, Shot Atoll, Spheres plus Irregulars)
Chain 99 (^{99}Mo)						
57	4.8	16.0	10.2[b]	–	7.2	2.5
88	4.9	10.6	8.9[b]	–	6.6	4.0
125	5.8	9.8	8.4[b]	–	6.2	4.7
177	6.0	12.5	9.0[c]	35	6.0	5.7
297	12.4	13.2	15.2[c]	100	4.8	4.5
594	11.9	21.3	5.7[c]	68	3.4	1.6
840	3.1	24.3	4.7[c]	58	–	–
Chain 89 (^{89}Sr)						
57	0.075	0.24	0.36[b]	–	0.086	0.063
88	0.065	0.17	0.28[b]	–	0.11	0.074
125	0.046	0.19	0.24[b]	–	0.12	0.082
177	0.042	0.14	0.18[c]	0.24	0.12	0.062
297	0.043	0.12	0.22[c]	0.26	0.13	0.044
594	0.044	0.11	0.11[c]	0.063	0.046	0.063
840	0.075	0.070	0.042[c]	0.031	–	–
Chain 140 (^{140}Ba)						
57	0.32	1.28	0.67[b]	–	0.20	0.25
88	0.27	0.74	0.54[b]	–	0.22	0.28
125	0.20	0.99	0.45[b]	–	0.22	0.30
177	0.17	0.77	0.39[c]	0.48	0.23	0.23
297	0.15	0.75	0.49[c]	0.74	0.25	0.18
594	0.16	0.67	0.31[c]	0.18	0.13	0.24
840	0.031	0.41	0.12[c]	0.085	–	–

[a] Derived from Morgenthau *et al.* (1960).
[b] Irregulars plus spheres, near ground zero.
[c] Irregulars only, spheres removed, near ground zero.

show a number of instances in which spheres arose by coalescence of heterogeneous droplets (Norman and Winchell, 1967).

The $1/D^2$ behavior observed for volatile isotope chains in particles in excess of 300 μ is explicable on two grounds. Vapor phase controlled diffusion may lead to an activity proportional to the diameter of the particle for a given exposure time and hence to an activity concentration inversely as the square of the diameter. Also, larger particles are falling out of the volatile-rich cloud faster than smaller particles, leading to a decrease in activity concentration with increasing diameter. It is of some interest also that the uptake of refractory chains proceeds as $1/D$, indicating incorporation as the area for these large particles. The refractory-containing droplets are probably attached very early (in the first few minutes); vapor phase controlled diffusion probably plays no role here.

The activity concentration maximum of a volatile chain is understandable in terms of residence time in the cloud. A 100-μ particle found in the prompt fallout pattern could not have originated from so high a position in the cloud as a 300–500-μ particle and hence would have been exposed to volatile chain vapors for a shorter time.

Attempts to achieve a basic understanding of fractionation behavior as a function of explosion conditions and yield have been made by a number of workers. The thermodynamic model of Miller (1960) was a major step in placing fallout studies on a scientific basis. Norman *et al.* (1970) have carried the Miller model forward with a number of significant refinements, both conceptual and experimental. The radial distribution model of Freiling (1961, 1963) leads to considerable systemization in the treatment of debris behavior. Heft (1970) has achieved considerable success in describing fractionated systems with a nonphenomenologi-

cal model that introduces a minimum number of basic assumptions.

Laboratory studies by Freiling (1970) and Adams *et al.* (1967a, 1967b) have recently focused on the kinetic aspects of the pickup of fission-product vapors by molten oxide droplets. Similar studies are under way in a number of other laboratories, using methods of high-temperature chemistry. A satisfactory physical model of debris formation processes must probably include due consideration of vaporization, condensation, and agglomerative processes. Equally important is a better understanding of the early thermal and mixing history within the ball of fire and developing nuclear cloud.

RELATIVE VOLATILITIES OF SOME FISSION PRODUCT CHAINS

It is appropriate to mention briefly fundamental chemical differences between subclasses of volatile fission product mass chains. Chain 89 is volatile mainly because of the 3.2-min ^{89}Kr; ^{89}Rb, another chain member, will behave as a volatile element during the initial stages of fireball cooling when only the most refractory elements have condensed. On the other hand, if the temperature is well below the boiling point of rubidium or its oxides, the latter will quickly attach to a debris particle surface. Similarly, the volatility of chain 131, usually measured as ^{131}I, is conferred by the relatively low boiling properties of Sn, Sb, and Te oxides. When the temperature has fallen to below 1000° C, these elements can be considered to be no longer volatile for the usual range of partial pressures.

It is apparent that radiochemical composition of the fallout must change proceeding downwind from ground zero, becoming more enriched in volatile relative to refractory chains.

The Special Case of Underwater Explosions

Only an insignificant fraction (less than 0.1 percent) of the total megaton equivalents of fission detonations (see Table 3) has resulted from underwater bursts. These events have the unique characteristic of being point source injections. The initial oceanic distribution is a function of the yield and depth of detonation. Based on limited test experience in oceanic situations, for shallow explosions, from one third to two thirds of the debris may be found in the mixed layer in a pool many hundreds of meters in radius and mixed to the existing thermocline (approximately 100 m). The history of such an injection as it was influenced by currents, turbulent diffusion, and decay, is depicted in Figure 3 as measured from an aircraft and in Figure 4 as measured by an *in situ* gamma probe (Riel, 1962).

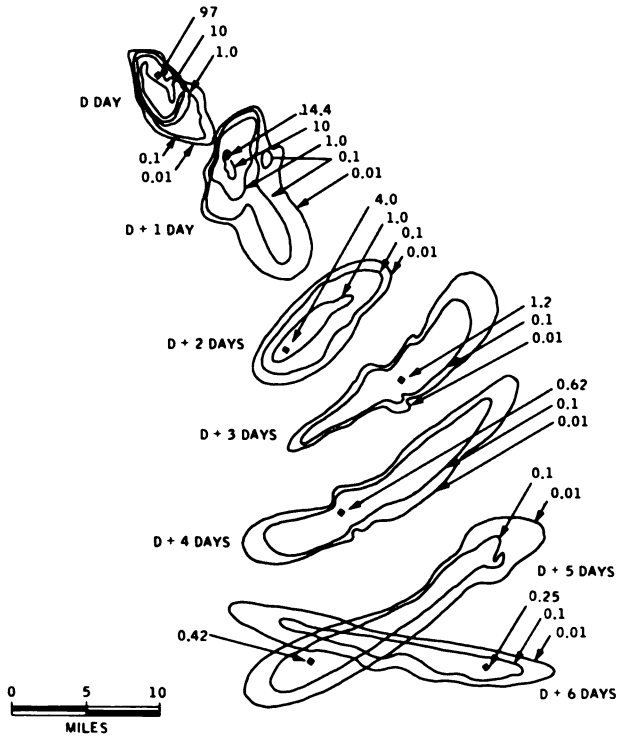

FIGURE 3 Surface pattern of distribution of radioactivity from a nominal-yield underwater explosion as measured from an aircraft at 500-ft (150-m) altitude. (Gamma contour values in mR/hr.)

Debris from deep explosions deposited in the thermocline layer will generally react to the same environmental influences; however, since the subsurface scale of turbulence is small, the predominant activity reduction mechanism is radioactive decay. Discrete lamina of radioactivity were observed in the thermocline layer after the WIGWAM test.* The fraction of the debris in these waters, as reported by Isaacs (1962), was estimated to be approximately two thirds of the total debris released by the explosion. Advection and diffusion of radioactivity in the ocean is discussed in detail in Chapter 4.

Fractionation of fission products created in underwater detonations is not severe (Freiling and Ballou, 1962). Individual fission product data from samples collected shortly after detonations indicated a variation of no more than 30 percent in predicted fission product ratios. An exception was ^{89}Sr from a deep burst, which varied by a factor of up to about 10 in relation to other fission product radionuclides.

*A 30-kt device detonated at a depth of 2,000 ft (610 m) in the Pacific at 29° N, 126° W, in 1955.

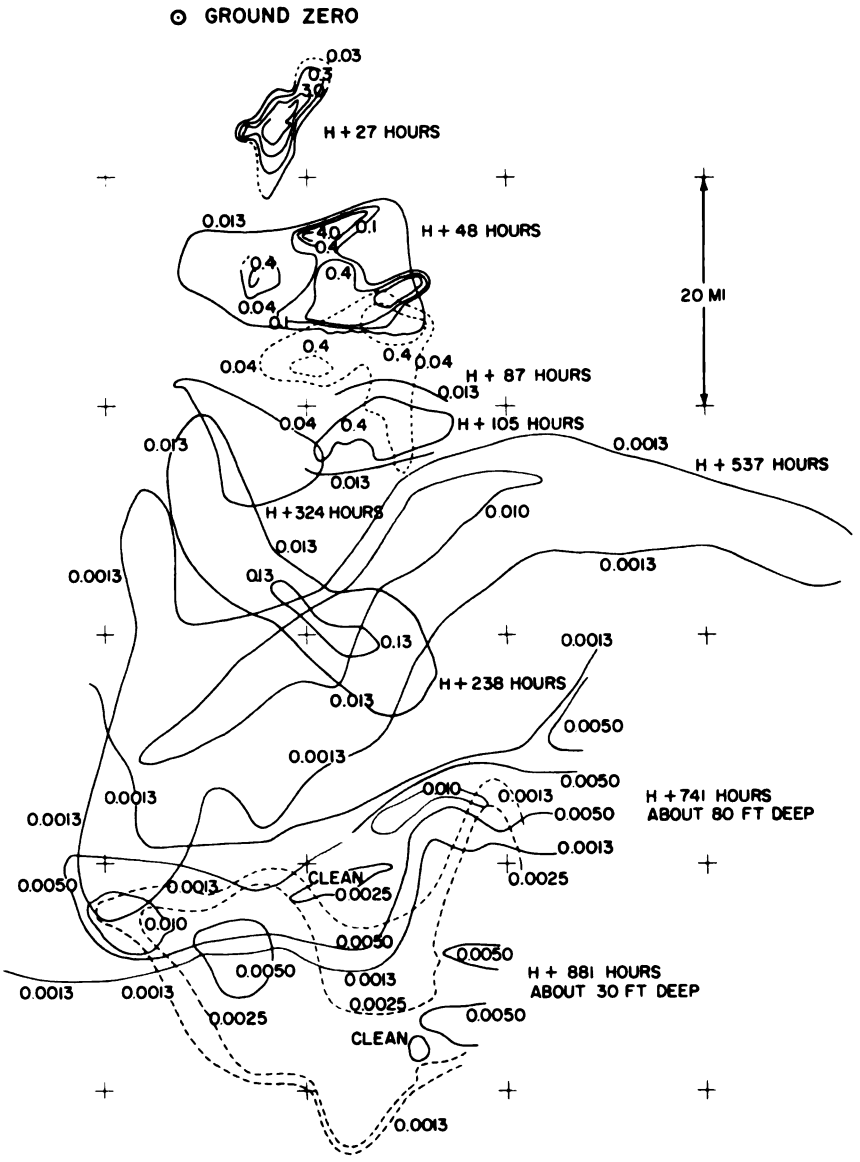

FIGURE 4 Subsurface pattern of distribution of radioactivity from a nominal-yield underwater explosion as measured by an *in situ* gamma probe (some lines are dashed for clarity of presentation). (Reprinted from Riel, 1962).

Nuclear Explosion Debris and Its Interaction with Seawater

The physical and chemical states of bomb debris in seawater are poorly understood. Some theoretical estimates have been made, a few determinations from actual debris have been reported, and some laboratory studies using stable element counterparts of detonation debris have been undertaken (Freiling and Ballou, 1962; Greendale and Ballou, 1954).

Radionuclide physicochemical states are largely determined by their state at the time of addition to the ocean (or by the initial state and the decay of their precursors) and by the effects of their chemical environment.

Little is known of the fundamental processes of formation of nuclear debris from an underwater explosion. In the absence of large quantities of bottom material, the initial processes can be visualized as forming debris consisting of vaporized and dissociated water, seawater salts, and device materials. For a 10-kt device detonated underwater, on the order of 7×10^6 kg of seawater will be vaporized. The bubble of debris will then contain, in addition to the vaporized device materials and the 7×10^6 kg of H_2O, 1.3×10^5 kg of chlorine, 7.7×10^4 kg of sodium and potassium, 1.1×10^4 kg of calcium and magnesium, 6.3×10^3 kg of sulfur, and less than 7×10^2 kg of any other seawater constituent. As this system cools, nucleation of the debris should be

governed by iron, calcium, and magnesium oxides. The initial nuclei would therefore be expected to consist of particles of calcium, magnesium, and iron oxides of less than 20 μ diameter. These would contain the refractory-like radionuclides. Upon further cooling, these, in turn, would serve as nuclei for deposition of more volatile radionuclides.

The release of fission products to seawater from nuclear debris depends primarily upon particle solubility and leaching properties. It has been established that the ease of leaching gross beta activity from fallout particles increases in the following order: tower-shot debris, silicate-burst debris, airburst debris. Some authors report that the solubility increases as the size of the debris decreases. The debris from coral surface bursts is reported to be highly soluble in seawater (Adams *et al.*, 1960).

For underwater explosions, release to the seawater would include late-condensing soluble fission products and colloidal particles bearing most of the activity. Due to the large surface-to-volume ratio of small particles, leaching of soluble radionuclides would occur with high efficiency, leaving the insoluble radionuclides in the colloidal state. It has been shown that the halides, alkali metals, and alkaline earth metals will generally occur as soluble species in their normal oxidation states. Certain other elements, such as B, P, Ce, As, Se, Mo, Tc, and Te, are expected to occur as oxygenated anions that would be found partially in solution and partially colloidal or particulate. Nearly all of the remaining elements may be expected to exist as oxides or hydrated oxides associated with colloidal or gross particulate matter. However, our understanding of the interaction of radionuclides in such a complex mixture of inorganic compounds and organic and biological material is far from complete.

As a result of limited experiments, Freiling and Ballou (1962) summarized the nature of underwater nuclear explosion debris in seawater. They found that the total activity in the soluble phase increased from 35 percent at one day after the event to approximately 60 percent after two weeks. This increase was attributed to the slow conversion of some of the constituent elements into soluble material, rather than to the production of new elements through radioactive decay.

The percentages of soluble and insoluble fractions of a number of radionuclides were also determined by Ballou (1963), as shown in Table 5. These data generally indicate greater solubility than that earlier reported by Greendale and Ballou (1954).

Similar investigations were carried out with waterborne debris from an underwater burst in which the fireball contacted the bottom of a coral lagoon. These data, after Ballou (1963), are reported in Table 6. Particle sizes ranged from 0.001 μ to 10 μ in diameter. The distribution of radionuclides in surface water was collected 2 hr after detonation and analyzed 31 hr after detonation. "Solid phase" refers to material remaining in the ultrafiltrate. Standard deviations were calculated from duplicate analyses. Deviation of sums from unity indicates a lack of mass balance or is an indication of sample contamination.

For the data in Table 6, radiochemical analysis for selected radionuclides was performed on duplicate aliquots of the gross samples, ultrafiltrates, and dissolved membranes. Results from successively separated fractions did not reveal any change in the physical state distribution of any radionuclide with time, but mass balance deficiencies prohibit firm conclusions in some cases. The results from successive ultrafiltrations have therefore been averaged. The results for each radionuclide discussed below are compared with the results of the two previous investigations. Included in each discussion is the mass balance achieved and the effect of an

TABLE 5 Percentages of Activity of Radionuclides in Ultrafiltrates from Surface Water Samples, Underwater Burst

Sample	Centrifugation Time (hr from zero time)	Relative Activity in Ultrafiltrate at Centrifugation Time (%)						
		^{95}Zr	^{99}Mo	^{239}Np	^{237}U	RE[a]	^{137}Te	^{140}Ba
1	1	35	64	–	54	33	0	35
	7	–	–	42	–	–	–	–
	19	43	66	44	80	19	0	–
2	7	–	–	41	–	–	–	–
	19	–	–	53	–	–	–	–
3	10	48	67	67	77	42	0	–
	19	–	–	52	–	–	–	–
	116	43	63	–	71	39	0	–
Average		42	65	50	71	34		35

[a]Gross rare earths and yttrium.

TABLE 6 Radionuclide Distributions 31 hr after Detonation in Lagoon Burst; H + 2 Surface Water Sample

Radionuclide	Fraction Distribution		
	Solid Phase	Colloidal Phase	Soluble Phase
^{89}Sr	0.00 ± 0.00	0.11 ± 0.01	0.99 ± 0.00
^{95}Zr	0.38 ± 0.01	0.16	0.03 ± 0.00
^{95}Nb	0.45 ± 0.00	0.39 ± 0.01	0.00 ± 0.00
^{99}Mo	0.28 ± 0.00	0.06 ± 0.01	0.60 ± 0.00
^{103}Ru	0.60	0.25 ± 0.00	–
^{132}Te	0.60 ± 0.00	0.18 ± 0.00	–
^{140}Ba	0.00 ± 0.00	0.01 ± 0.01	0.99 ± 0.01
TRE[a]	0.83 ± 0.02	0.14 ± 0.06	0.03 ± 0.00
^{237}U	0.02 ± 0.00	0.11 ± 0.01	0.04 ± 0.06
^{239}Np	0.47 ± 0.00	0.46 ± 0.01	0.02 ± 0.00

[a]Total rare earths.

additional membrane on the physical state distribution and on the mass balance observed. Mass balance deficiency may be taken as a measure of contamination potential, since it presumably arises from adherence of the radioactive species to the walls of the polyethylene container or to the surfaces of the ultrafilter apparatus.

^{89}Sr An average value of 4 ± 1 percent of the ^{89}Sr was found to be in the insoluble phase, and an average mass balance of 116 ± 5 percent was obtained. The results show a somewhat greater solubility than that found in underwater arcing experiments, but not outside the precision of the methods. The results were unaffected by the presence of a second membrane.

^{95}Zr and ^{95}Nb Analyses were performed for both of these radionuclides, and no distinction between their behavior was evident. The presence of a second membrane increased both the mass balance and the percentage of activity appearing in the colloidal fraction. With either a single or double membrane, 0–8 percent of the activity was found in the soluble state, in fair agreement with underwater arcing results. With only a single membrane, an average of 48 percent was found to be insoluble, but the results were spread from 34 to 66 percent. The results from the deep underwater burst are within this range.

^{99}Mo Mass balance was consistently good, averaging 102 ± 5 percent. Using a single membrane, an average value of 68 ± 5 percent was found to be soluble, in good agreement with the deep underwater burst results. With a double membrane, 26 ± 5 percent was found to be soluble, in agreement with underwater arcing results.

^{103}Ru It was found that 82 ± 5 percent of the ruthenium was insoluble, the mass balance being 105 ± 6 percent. No double-membrane effect was noted. The solubility is therefore greater than indicated by the value of 1 percent from the underwater arcing experiments.

^{132}Te The insoluble fraction was found to contain 64 ± 9 percent of the activity and the mass balance was 81 ± 12 percent. No double-membrane effect was noted. These results are in better agreement with those from underwater arcing than with those from the deeper underwater burst.

^{140}Ba This radionuclide showed a much higher solubility than the single determination made for the deep underwater burst would indicate. Only 5 ± 1 percent was found in the insoluble fraction, the mass balance being 88 ± 4 percent. No double-membrane effect was noted. The results are similar to those obtained for ^{89}Sr.

Total rare earths The insoluble fraction contained 67 ± 3 percent of the activity, in accord with the deep underwater burst results, but the mass balance was only 71 ± 3 percent. No double-membrane effect was noted. Cerium-141, 144 was found to behave similarly. This behavior is not inconsistent with the results from the underwater arcing experiment, in which it was found to be soluble to the extent of 3 percent or less.

^{237}U The insoluble fraction contained 46 ± 5 percent of the activity, but the percentage of activity found in the soluble fraction was quite variable. The deep underwater burst results also indicate considerable variability. No double-membrane effect was noted.

^{239}Np With single membranes, 50 ± 2 percent of the activity was found to be insoluble, in agreement with the deep underwater burst results. With a double membrane, however, 93 percent was found to be insoluble. As in the case of ^{237}U, the percentage of activity found in the soluble fraction was quite variable.

Radioactive Fallout from Nuclear Weapons Tests

The specific character of the oceanic contamination from nuclear explosions depends very directly upon the nature, size, and physical location of the event. Thus, underwater bursts introduce radioactive contamination directly and immediately into the sea in a relatively small and well-defined area. Surface or air detonations, depending upon the size of the burst, distribute the debris in the atmosphere to varying altitudes and horizontal distances. The extent of these atmospheric dispersions directly influences the rate of fallout of the radioactive material and its ultimate distribution on the earth's surface.

The bulk of the nuclear debris from large surface or air bursts enters the stratosphere. Once in the stratosphere, this material is, for practical purposes, insulated from the earth's weather, and the rate and extent of its distribution are determined by meteorological processes in the stratosphere. In general, the stratospheric mixing rate is more rapid within the hemisphere of injection than either the fallout rate or the rate of intrahemispheric mixing. These factors result in a generally characteristic pattern of distribution of the radioactive aerosol within that hemisphere.

Removal of radionuclides from the stratosphere occurs by downward mixing and diffusion, with the major entry into the troposphere occurring in the middle and upper latitudes during late winter and early spring, in both hemispheres. The rate of stratospheric depletion of nuclear debris depends, to a degree, upon the latitude of the injection and the altitude of stabilization of the particles.

This rate, termed the "stratospheric half residence time" has been empirically observed to resemble a simple exponential decay. The minimum rate appears to be about 6 months, for nuclear tests in the lower stratosphere in polar regions. Debris injected into very high altitudes may take 2 years or longer before reaching the ground. It is interesting, however, that since the partial test ban treaty went into effect at the end of 1962, the stratospheric half residence time has remained quite constant at about 10 months.

The rate of intrahemispheric mixing in the stratosphere is slow, compared to the average observed stratospheric fallout rate; hence, the bulk of the nuclear debris remains within the hemisphere of introduction. Upon leaving the stratosphere, the debris is deposited on the earth's surface primarily as a direct result of precipitation and with a "tropospheric half residence time" of approximately 30 days or less.

The seasonal release of stratospheric debris into the troposphere, the probable latitudinal distribution of this release, and the indicated precipitation patterns on the ground combine to produce the observed spring maximum in fallout and global distribution with higher values in the mid-latitudes. Figure 5 illustrates a typical latitudinal distribution of stratospheric fallout.

Of the many radionuclides produced in the testing of nuclear weapons, ^{90}Sr has been examined the most intensively and extensively because its relatively high fission yield (~3.5 atoms per 100 fissions) and its bone-seeking character and long physical half-life make it potentially the greatest fallout hazard to man. Measurements of ^{90}Sr in a variety of environmental and biological media, including human specimens, have been made since the beginning of weapons testing. Atmospheric measurements coupled with the direct assay of ^{90}Sr in soil have made possible the assembly of a ^{90}Sr inventory. Interestingly enough, the inventory of ^{90}Sr in the atmosphere (mainly in the stratosphere) plus that present on the surface matches quite closely the total production of ^{90}Sr calculated from weapons testing to date. This does not imply detailed knowledge of the precise ^{90}Sr yield, or indeed the explosive yield, of each and every detonation, but it does apply to the time average of such events. Hence, we may conclude that we know the production and distribution (on a broad scale) of this particular radionuclide. The annual production and rate of deposition of ^{90}Sr are indicated in Table 7.

Table 8 lists the annual deposition of ^{90}Sr by 10° latitude bands for the period 1958–1966. These values were computed from the data of the worldwide sampling network and are based upon the assumption that there is no systematic difference in fallout efficiency between ocean and land surfaces. With that restriction, the information in Table 8 may be utilized as a direct measure of input to the oceans as a function of time.

Table 8 may also be used to derive stratospheric deposition values for other fission product nuclides on the further assumption that the ratio between such nuclides and ^{90}Sr is constant. There is substantial evidence that this can be assumed with some confidence for ^{137}Cs; hence, the ^{90}Sr values of Table 8 can be converted to ^{137}Cs values by multiplying by the constant 1.5. Calculation of amounts of other relatively long-lived fission products, such as ^{144}Ce and ^{95}Zr, from ^{90}Sr ratios should be used with caution and must be

TABLE 7 Worldwide Production and Deposition of ^{90}Sr (in MCi)

Year	^{90}Sr Produced	^{90}Sr Deposited
1945–1958	9.1	5.6
1959	0	1.1
1960	0	0.4
1961	2.5	0.4
1962	7.6	1.6
1963	0	2.6
1964	0	1.9
1965	0	1.0
1966	0	0.4

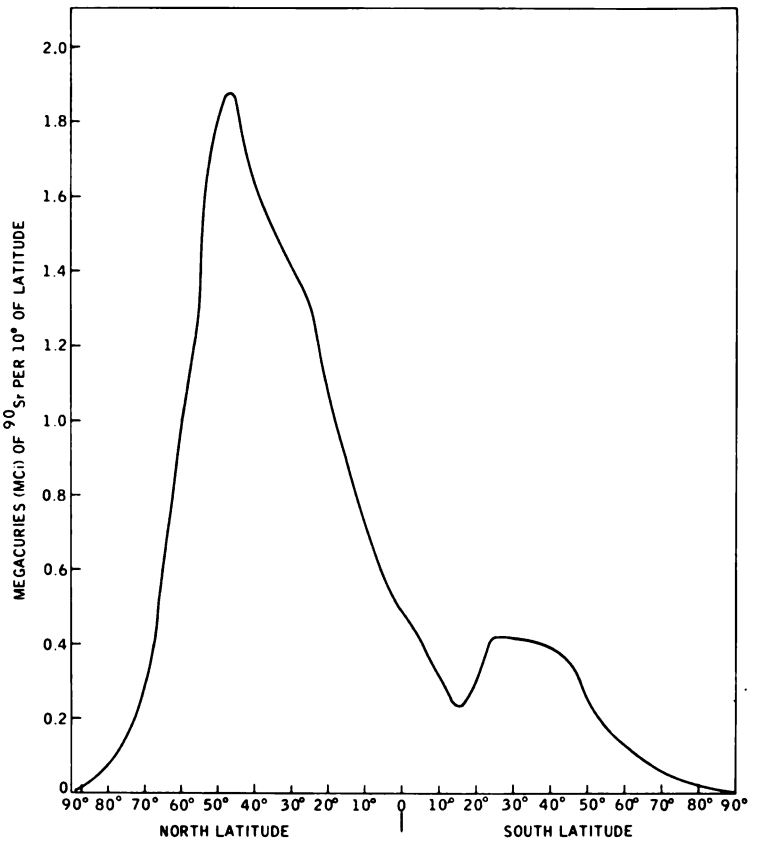

FIGURE 5 Latitudinal distribution of ^{90}Sr fallout (1958–1967).

corrected for radioactive decay, since their half-lives are relatively short.

A calculation of the distribution of ^{55}Fe produced as a product of activation by neutrons presumably on materials of the nuclear devices also was made based upon the $^{55}Fe/^{90}Sr$ ratio. A very substantial amount of this nuclide was made and injected into the stratosphere during the U.S. and Soviet test series in 1961 and 1962. The fallout data for ^{55}Fe, though sparse, suggest that it had apparently become rather well mixed with the ^{90}Sr in the stratosphere by 1963. On this basis, an average ratio of ^{55}Fe to ^{90}Sr of 9.4 was derived and used for the compilation of Table 9.

Future Applications of Nuclear Explosives

Some potential peaceful applications of nuclear explosives constitute a potential source for the introduction of radionuclides into the marine environment. The proposed applications that could contribute the most radioactivity to the oceans involve the use of nuclear explosives for the construction of harbors or canals.

The U.S. Atomic Energy Commission's Plowshare Program for developing potential uses for nuclear explosives is in its early developmental stages. While some experiments have been conducted in terrestrial situations, none has been attempted in the marine environment. Guidelines for the employment of nuclear explosives and for the design of the explosives themselves are still being developed and refined.

Environmental contamination resulting from nuclear excavations would differ significantly in at least three ways from that resulting from atmospheric weapons tests. First, most of the radioactivity produced would be retained in the broken rock that falls back into the crater and in the ejecta in the immediate vicinity of the detonation site, and any portion that moves from the detonation site will do so only by surface runoff or groundwater transport. Second, the radioactivity produced per megaton of explosive force will be substantially reduced by employing explosives with low fission yields, by using nonactivating neutron absorbers to reduce the neutron activation of components of the explosive and of the soil, and by using explosive components whose activation products are of low biological significance. Finally, because low-fission-yield devices will be employed, activation products will equal or surpass fission products in curies produced and in biological significance. In this respect, however, it is important to note that, of the radionuclides thus far introduced into marine or freshwater environments, the greatest potential hazard to man has involved

TABLE 8 Annual Deposition of ^{90}Sr Since 1958, by 10° Bands of Latitude (in MCi)

Latitude	1958	1959	1960	1961	1962	1963	1964	1965	1966	Total
80°-90° N	0.003	0.002	0.001	0.001	0.002	0.007	0.002	0.001	0	0.019
70°-80°	0.025	0.016	0.006	0.006	0.017	0.047	0.014	0.008	0.003	0.142
60°-70°	0.064	0.059	0.011	0.020	0.089	0.200	0.087	0.030	0.012	0.572
50°-60°	0.114	0.157	0.031	0.044	0.168	0.376	0.262	0.109	0.042	1.293
40°-50°	0.153	0.206	0.043	0.065	0.285	0.540	0.354	0.149	0.056	1.851
30°-40°	0.147	0.218	0.043	0.064	0.247	0.358	0.248	0.137	0.051	1.513
20°-30°	0.106	0.192	0.038	0.045	0.214	0.358	0.239	0.091	0.034	1.317
10°-20°	0.024	0.104	0.035	0.035	0.145	0.250	0.180	0.094	0.036	0.903
0°-10°	0.019	0.021	0.025	0.031	0.113	0.184	0.093	0.069	0.026	0.581
0°-10° S	0.044	0.034	0.024	0.021	0.096	0.063	0.073	0.039	0.015	0.409
10°-20°	0.041	0.023	0.018	0.017	0.028	0.033	0.036	0.028	0.011	0.235
20°-30°	0.055	0.031	0.027	0.044	0.048	0.056	0.068	0.062	0.024	0.415
30°-40°	0.067	0.032	0.022	0.033	0.032	0.050	0.075	0.068	0.026	0.405
40°-50°	0.030	0.022	0.028	0.024	0.036	0.049	0.076	0.071	0.027	0.363
50°-60°	0.018	0.015	0.017	0.015	0.017	0.023	0.032	0.026	0.010	0.173
60°-70°	0.010	0.007	0.008	0.008	0.009	0.012	0.016	0.012	0.005	0.087
70°-80°	0.004	0.002	0.003	0.003	0.003	0.004	0.005	0.005	0.002	0.031
80°-90°	0	0	0	0	0	0	0.001	0.001	0	0.002
Total	0.924	1.141	0.380	0.476	1.549	2.610	1.851	1.000	0.380	10.311

TABLE 9 Deposition of ^{55}Fe from Weapons Tests, by 10° Bands of Latitude (in MCi of ^{55}Fe, decay corrected to October 15, 1961)

Latitude	1962	1963	1964	1965	1966	Future[a]	Totals
80°-90° N	0.01	0.06	0.01	0.01	0	0	0.09
70°-80°	0.08	0.44	0.13	0.07	0.03	0.01	0.76
60°-70°	0.42	1.88	0.82	0.28	0.11	0.04	3.55
50°-60°	0.79	3.54	2.36	1.02	0.39	0.16	8.26
40°-50°	1.34	5.10	3.32	1.40	0.53	0.20	11.89
30°-40°	1.16	3.36	2.32	1.29	0.48	0.18	8.79
20°-30°	1.00	3.36	2.33	0.85	0.32	0.12	7.88
10°-20°	0.68	2.34	1.68	0.89	0.34	0.13	6.06
0°-10°	0.53	1.72	0.87	0.65	0.25	0.09	4.11
Total, northern hemisphere	6.01	21.80	13.74	6.46	2.45	0.93	51.39
0°-10° S	0.45	0.59	0.68	0.36	0.14	0.09	2.31
10°-20°	0.13	0.31	0.34	0.26	0.10	0.13	1.27
20°-30°	0.22	0.52	0.64	0.59	0.23	0.12	2.32
30°-40°	0.15	0.47	0.70	0.64	0.25	0.18	2.39
40°-50°	0.17	0.46	0.72	0.66	0.25	0.20	2.46
50°-60°	0.08	0.21	0.30	0.25	0.10	0.16	1.10
60°-70°	0.04	0.11	0.15	0.12	0.05	0.04	0.51
70°-80°	0.01	0.04	0.05	0.05	0.02	0.01	0.18
80°-90°	0	0	0.01	0.01	0	0	0.02
Total, southern hemisphere	1.25	2.71	3.59	2.94	1.14	0.93	12.56
Total, world	7.26	24.51	17.33	9.40	3.59	1.86	63.95

[a]Based on Stardust 0.2 MCi ^{90}Sr from pre-1963 tests. This material was evenly distributed over the northern and southern hemispheres; thus, fallout is assumed symmetrical. The ratio of ^{55}Fe to ^{90}Sr changes with time; using a constant ratio of 9.4 for the period 1962–1966 introduces an error.

activation products such as ^{32}P, ^{65}Zn, and ^{55}Fe. In underground explosions, ^{3}H contamination of groundwater may be a biological hazard, depending on the quantity produced.

There are three sources of the radionuclides produced in a thermonuclear cratering explosion: fission products, radionuclides induced in the device materials, and radionuclides induced in the environmental media surrounding the device. In addition, between 7×10^6 and 5×10^7 Ci of tritium, depending upon the nature of the thermonuclear reaction, may be produced.

In a thermonuclear explosion, roughly 10^{27} neutrons will be produced per megaton of yield. In addition, a large number of protons, deuterons, tritons, helium-e nucleii, and alpha-emitting radionuclides will be generated. These neutrons and charged particles will react with the materials of the device to produce a wide variety of radionuclides.

Estimation of the activation radionuclides produced in a nuclear event is difficult because many of the cross-section values necessary for the calculations are unknown. However, using the data that are known, such as Ng's calculations of the activation products in major rock types (1965), and using worst-case assumptions where needed, estimates of the potential biological hazard of a device can be made (Kaye *et al.*, 1969; Tamplin, 1967; Ng and Thompson, 1966; Burton and Pratt, 1967; Tamplin *et al.*, 1968; Martin, 1969).

When an explosive is detonated on a tower, all of the radioactivity produced is released to the atmosphere, but when the explosive is detonated underground, only a fraction of the activity is released to the atmosphere. When the explosive is buried deep enough, all of the activity remains underground. In cratering experiments, in which the expanding cavity ruptures the surface to form the crater, the material above the explosive acts like a filter bed and removes a large portion of the radioactive material that would otherwise be released to the atmosphere. The efficiency with which a radionuclide is removed depends on its chemical and physical properties or on those of its precursors during the venting process. Fission products such as ^{137}Cs and ^{90}Sr, which have rare-gas precursors during the venting process, are found in much higher relative concentrations in the cloud than are refractory radionuclides that do not have gaseous precursors, such as ^{95}Zr or ^{147}Nd. Therefore, to the extent that it is possible, the fraction released to the atmosphere must be determined for each specific radionuclide of interest.

The final physical and chemical forms of the radionuclides produced in a nuclear cratering explosion are poorly known, except for a few species. The final forms depend, as in the case of fractionation, upon the chemical nature of the species present during cavity expansion and venting. The chemical species present depend upon such factors as the water content of the medium and the overall oxidation-reduction potentials of the mixture. Large quantities of rock are melted and vaporized along with the materials of the device itself. As the cavity expands, the vaporized materials begin to condense, the order of condensation depending upon the volatilities of the materials. It is possible to alter this overall process by placing in the immediate vicinity of the device materials that could, for example, change the overall oxidation-reduction potential of the mixture and, therefore, the chemical species present. Whether such a procedure should be considered, of course, depends upon the associated biological hazard.

The U.S. Atomic Energy Commission (1967c) has released the following information concerning the radioactivity released to the atmosphere by cratering explosives:

> In order to plan for major excavation projects, the following factors relative to release of radioactive debris should be taken into account: The amount of radioactivity airborne in the cloud and in the fallout is minimized by scavenging during the venting process, by special emplacement techniques, by utilizing minimum fission explosives, and by employing extensive neutron shielding. Based on reasonable assumptions about these factors, the following information can be used in planning for cratering events of useful magnitude. For each individual nuclear explosive detonated, the sum of fission products airborne in the radioactive cloud and in the fallout can be expected to be as low as the equivalent of 20 tons. The tritium release may be less than 20 kilocuries per kiloton of total yield. The sum of activation products airborne in the radioactive cloud and in the fallout may be expected to be as low as the amounts shown in the following table:

Representative Set of Induced Radioactivities at Detonation Time (Total in Cloud and Fallout)

Nuclide	Nuclide Production, Kilocurie for Yield of		
	100 KT	1 MT	10 MT
^{24}Na	200	800	2,000
^{32}P	0.1	0.4	0.8
^{45}Ca	0.01	0.03	0.06
^{54}Mn	0.1	0.3	0.7
^{56}Mn	6,000	20,000	50,000
^{55}Fe	0.04	0.15	0.3
^{59}Fe	0.04	0.15	0.3
^{185}W	6	10	14
^{187}W	300	500	700
^{203}Pb	1,000	7,000	20,000
Other	15	20	40

NUCLEAR REACTORS

Nuclear reactors have been designed and built for a variety of purposes, as indicated by Table 1. Each purpose involves a design concept keyed to the desired form of energy output, which in turn depends on the intended application. Reactors are designed to produce power, neutrons, radio-

and several tests of this technique were conducted (Nuclear Science and Engineering Co., 1958). Since then, the technology of fluorometers has advanced considerably, and it is now possible to measure dye concentrations of the same low order as radioisotopes—0.5 parts per billion. Dyes largely have superseded isotopes in the measurement of water mixing processes. Selected radioisotopes such as ^{65}Zn and ^{32}P continue to be useful for studies of ecological systems (Pomeroy and Odum, 1966; Hooper and Ball, 1966).

SUMMARY

Radioactivity in the oceans can result from numerous sources. Natural atmospheric and geologic processes issue discrete radionuclides in certain forms and distributions; these are briefly described. Man's uses of nuclear energy result in another, wider spectrum of radionuclides of varying characteristics. The biological distribution in the ocean of man-made radionuclides depends greatly on the initial physicochemical form of the radioactivity, and this, in turn, depends on the particular use or application of nuclear energy. Nuclear energy applications are described, indicating the particular radionuclides involved in each application and the form of the radioactivity likely to be introduced into the ocean. The applications include nuclear explosions at various positions relative to the earth's surface; nuclear reactors for electric power, fuel production, ship propulsion, and space vehicle propulsion; nuclear fuel processing; waste disposal; auxiliary power generators; and radioisotopic tracers.

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TABLE 1 Radiation Dose Rates in mrad/Week in Various Tissues of Cod (*Gadus callarias*) and Haddock (*Gadus aeglefinus*), from the Decay of ^{40}K and ^{226}Ra[a]

Radionuclide		Cod				Haddock			
		Muscle	Skin	Bone	Fins	Muscle	Skin	Bone	Fins
^{40}K	β	0.43	0.25	0.28	0.16	0.36	0.25	0.32	0.16
	γ	0.03	0.01	0.01	0.01	0.03	0.01	0.02	0.01
^{226}Ra		–	0.020	–	–	–	0.040	–	–

[a]After Fedorov (1965).

TABLE 2 Estimates of Natural-Radiation Dose Rates to Plaice (*Pleuronectes platessa*) in the Irish Sea[a]

Source	Type of Radiation	Dose in mR/week
Natural activity in seawater (^{40}K only)	γ	0.07
	β	0.18
Natural activity in seabed (estimate)	$\beta\gamma$	1.00
Cosmic radiation at 20 m		0.10
^{40}K in fish	γ	0.01
	β	0.22
Total		1.58 mR/week or 0.082 R/year

[a]Data from Woodhead (personal communication).

absorbed by organisms or adsorbed on their surfaces; these nuclides will contribute to the internally delivered dose to a greater or lesser extent, depending on the size and complexity of the organism and the energy of the emission. In addition, any of the parameters discussed in Chapter 7 or any group of them influences the exposure of organisms to radiation.

Laboratory studies of radiation effects on individuals have been made with a variety of techniques. The basic studies of sensitivity to acute doses of radiation have mainly employed x rays, while studies attempting to reflect the environmental conditions have used ^{60}Co and ^{137}Cs sources or radionuclides in solution. Environmental and population studies have been more limited, however.

Our intention in this chapter is to consider only that research pertinent to the prediction of what might occur in the marine environment; therefore, we have not considered all of the studies that have been carried out using marine organisms in radiobiological research.

NATURAL RADIOACTIVITY

The sources contributing to the natural background radiation dose of organisms in the marine environment are cosmic rays and the natural radioactivity in the earth's crust, present in seawater sediments and biota (Folsom and Harley, 1957). More than 60 radionuclides have been identified within the marine ecosystem (Chapter 2), and, based on these measurements, some calculations have been made of the dose rates to which the biota is exposed.

Cherry (1964), in studies with phytoplankton from the open sea, shows that dose rates from total alpha activity in organisms could range from 230 mR/yr to 2.8 R/yr. Fedorov (1965) has calculated the tissue dose from ^{40}K and ^{226}Ra in cod, *Gadus callarias*, and in haddock, *Gadus aeglefinus* (Table 1). Estimates have been made (Ministry of Agriculture, Fisheries and Food, 1967) of the beta and gamma dose rate from the environment to the predominantly bottom-living plaice, *Pleuronectes platessa*, in the Irish Sea (Table 2). The major source of radiation as far as this species of fish is concerned is the seabed.

Recent measurements of ^{210}Po in marine organisms suggest that radiation from natural alpha emitters contributes significantly to the radiation dose to organisms (Beasley, 1968).

MORTALITY INDUCED BY ACUTE RADIATION EXPOSURE

Lethal amounts of acute radiation differ widely among organisms because of biological variations related to such factors as species, age, physiological state, and body size. In the aquatic environment, these variations are further complicated by the interaction of environmental factors such as temperature, dissolved oxygen, chemical composition, and salinity. Nevertheless, exclusive of the eggs and larvae of invertebrates and fish, most of the freshwater and marine organisms for which data exist are relatively radioresistant.

Marine species differ little in radiation tolerances from freshwater species.

Values of LD_{50} (lethal dose resulting in 50 percent mortality) for acute irradiation of aquatic organisms have been listed by Donaldson and Foster (1957) and by Polikarpov (1966). In general, there is a relationship between radioresistance and the phylogeny and ontogeny of the organism. Primitive forms are more resistant than the complex vertebrates, and older organisms are more resistant than the young. Bacteria and algae may tolerate doses of thousands of roentgens, but freshwater fish, the most sensitive group listed by Donaldson and Foster (1957), were affected by considerably lower doses. The LD_{50} for adult rainbow trout, *Salmo gairdneri*, ranged from 300 to 3,000 R; and for the most sensitive stage of a developing trout egg, the LD_{50} value was as low as 16 R.

Despite research into lethal effects of radiation for over 50 years, surprisingly few LD_{50} values have been determined for marine organisms. Polikarpov (1966) lists LD_{50} values for 50 species of aquatic organisms, but the majority of these are freshwater organisms. White and Angelovic (1967) provided some LD_{50} values for several marine species irradiated with a ^{60}Co source. For six species of marine adult fish, the $LD_{50/30}$ (lethal dose for 50 percent mortality in 30 days) ranged from 1,050 R to 5,550 R, similar to values for freshwater fishes.

White and Angelovic (1965, 1966) point to the need to describe radiation tolerances in terms of time curves for mean lethal dose rather than in terms of the usual $LD_{50/30}$. Figures 1 and 2 show the time curves for mean lethal dose for 14 marine fishes and invertebrates. Figure 2 shows, for example, that for the first 25 days, the oyster *Crassostrea virginica* is much more resistant to radiation than the clam *Mercenaria mercenaria*; after 80 days, however, the LD_{50} for the clam was greater than for the oyster.

LD_{50} curves are needed, especially for the dominant economically important marine species in different ecosystems. However, because the levels of radiation required to kill marine organisms are so high, actual kills by radiation in the environment are extremely unlikely. These experimental LD_{50} curves will, however, keep in proper perspective our view of the likelihood of mortality in marine organisms from acute radiation.

CHRONIC EXPOSURE

External Radiation

Donaldson and Bonham (1964, 1966) have taken advantage of the migratory habit and the fecundity of chinook salmon, *Oncorhynchus tshawystcha*, to make a continuing long-term study of the effect on a population of chronic low-level irradiation from a ^{60}Co source during embryonic development. Eggs were first irradiated at 0.5 R/day from shortly

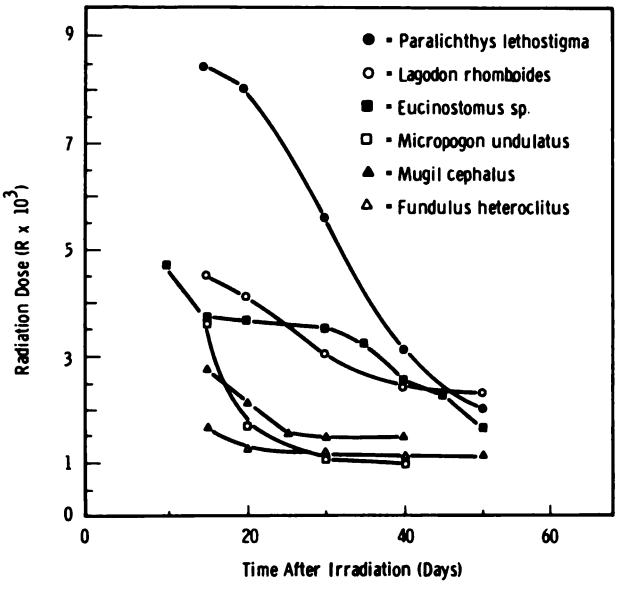

FIGURE 1 Mean lethal dose time curves for several species of marine vertebrates showing the dose-time combination at which 50 percent of the experimental animals died. (Reprinted with permission from White and Angelovic, 1966.)

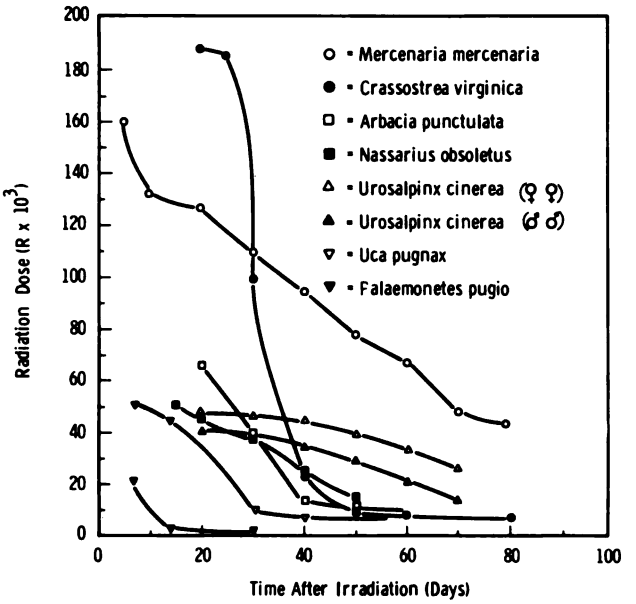

FIGURE 2 Mean lethal dose time curves for several species of marine invertebrates showing the dose-time combinations at which 50 percent of the experimental animals died. (Reprinted with permission from White and Angelovic, 1966.)

after fertilization until feeding commenced. The total dose was 33–40 R. The fingerlings were reared and then allowed to migrate to sea. Those that returned to the hatchery during the second year were precocious males; during the third and fourth years following irradiation, both male and female adults returned. Various crosses were made, some of which were reirradiated at 1.3 R/day to a total dose of 95 R. The initial dose chosen was about 40 times the calculated maximum dose that the germ cells and young salmon could receive in the Columbia River before migration to the sea. The radiation level used in the experiment was appreciable, amounting to 10^3 times the normal background, or 0.02 mR/hr.

These series of long-term experiments involving large numbers of organisms (96,000 to 256,000 fingerlings were released per experiment) indicate that irradiation at 0.5 R/day from the fertilization stage to the feeding stage produced no damage to the stock sufficient to reduce the reproductive capability over a period of slightly more than one generation. Although abnormalities in young fish were increased by irradiation, the number of adults returning was not affected. On the contrary, the irradiated stock returned in greater numbers and produced a greater total of viable eggs than the control stock.

Brown and Templeton (1964) and Templeton (1966) report on a series of experiments in which the eggs of plaice, *P. platessa*, were irradiated with a ^{137}Cs source. Total doses ranging from 0.6 to 500 R were used, at rates of 10 mR/hr to 1 R/hr from fertilization until hatching. No significant differences at hatching were observed in the survival or in the number of abnormal larvae produced.

Engel (1967) reports on studies of the effects of chronic low-level irradiation on the growth and survival of young blue crabs (*Callinectes spidus*). Single acute exposures had indicated that the sensitivity of these crabs is similar to that observed for other marine invertebrates. In the chronic irradiation experiment, the crabs were irradiated at an average of 22.5 hr/day at rates of 3.2, 7.3, or 29.0 rads/hr. The total radiation doses received over 70 days were 5,105, 11,502 and 45,693 rads. A significant number of deaths due to radiation occurred only among the crabs that received the highest radiation dose. The death rate of the crabs that received 3.2 and 7.3 rads/hr was similar to that of the controls. All irradiated and control crabs molted at least once. The numbers of second and third molts were affected by the radiation dose. The crabs that were irradiated at 29.0 rads/hr molted least, and none had three successful molts. Crabs that received 3.2 and 7.3 rads/hr underwent more second and third molts than did the controls, although this difference was not significant.

Radionuclides in the medium do not constitute the only radiation source to which pelagic organisms are exposed in the environment. A high degree of sorption of radionuclides into or onto the egg, for example, could give rise to a radiation dose within, or in the immediate vicinity of, the developing embryo that would be greater than that arising from the medium alone (Polikarpov and Ivanov, 1961, 1962; V. M. Brown, 1962; Brown and Templeton, 1964; Polikarpov, 1966). In addition, radiation from radionuclides incorporated into the developing tissues of an embryo may also be more effective in causing damage than external radiation alone (Polikarpov, 1966).

Hibiya and Yagi (1956) and Mikami *et al.* (1956), using fallout ash and rainwater residues from weapons tests, reported on the effects of these materials on the development of fish eggs. Concentrations in excess of 2×10^{-9} Ci/liter were found to be lethal, and abnormalities and delay in hatching were observed in concentrations down to 4×10^{-10} Ci/liter. The interpretation of these data must remain doubtful, however, since the radionuclide concentrations and chemical compositions of the ash and residues were not determined.

Polikarpov and Ivanov (1961, 1962), Ivanov (1965), and Polikarpov (1966) reported on the effect of ^{90}Sr-^{90}Y at low concentrations in seawater on the development of eggs of Black Sea fishes. Polikarpov and his co-workers, who pioneered the studies in this field, have reported on extensive studies with eggs of a large number of marine and freshwater species over the concentration range 10^{-14} to 10^{-4} Ci/liter of ^{90}Sr-^{90}Y. Reduced hatching of the larvae and early mortality were seen at concentrations of 10^{-7} Ci/liter and above, and the number of abnormalities increased significantly and with remarkable consistency at concentrations of 10^{-10} Ci/liter and above (Figure 3).

V. M. Brown (1962), Brown and Templeton (1964), and Templeton (1966) conducted similar experiments using eggs of the brown trout (*Salmo trutta*) and of plaice (*Pleuronectes platessa*), maintained from immediately after fertilization until hatching in water contaminated with ^{90}Sr-^{90}Y over the concentration range 10^{-10} Ci/liter to 10^{-4} Ci/liter. They did not observe any significant increase in mortality or in the production of abnormal larvae. Fedorov *et al.* (1964) reported that the eggs of plaice from the Barents Sea were sensitive to low concentrations of ^{90}Sr-^{90}Y in seawater. White and Angelovic (1966) report on the effects of chronic exposure to low levels of ^{137}Cs on the developing eggs and larvae of mummichogs (*Fundulus heteroclitus*). The concentrations of ^{137}Cs used, 3×10^{-7}, 3×10^{-6}, and 3×10^{-5} Ci/liter, produced no visible abnormalities. On the fourth day after fertilization, however, a general retardation in the rate of development was evident for those subjected to the highest concentration levels. On the nineteenth day after fertilization (approximately 50 percent hatch), the groups in the highest and the lowest concentrations had a slight reduction in the number of fish hatching, but this difference was no longer observable by the time hatching was completed.

Neustroev and Podymakhin (1966a), in similar studies with the eggs of the Atlantic salmon, *Salmo salar*, found

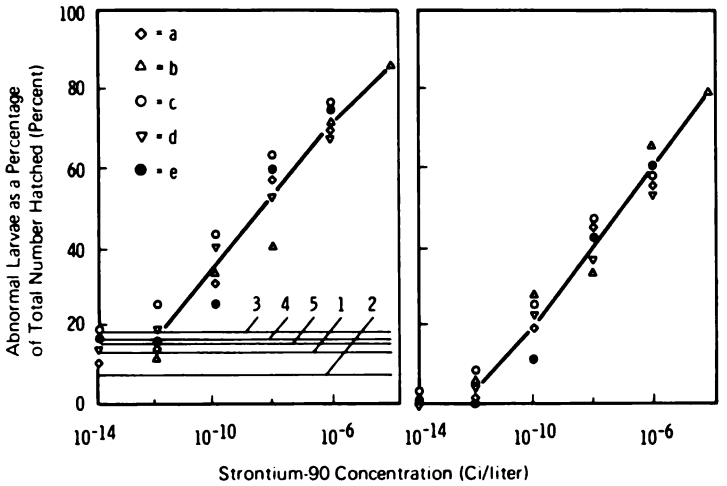

FIGURE 3 Dependence of proportion of abnormal larvae of Black Sea Fishes on strontium-90 concentration in surrounding seawater. (1) and (a) = abnormal mullet larvae; (2) and (b) = green wrasse (Labridae); (3) and (c) = horsemackerel; (4) and (d) = anchovies; (5) and (e) = from a mixture of pelagic eggs. (The figures denote the proportion of abnormalities in controls of the various species of fishes.) Left-hand graph without allowance for control, right-hand graph with allowance for control. (Reprinted with permission from Polikarpov, 1966.)

that at 10^{-10} Ci/liter ^{90}Sr-^{90}Y, the rate of development of the egg was the same as the control up to the stage of one-third development of the yolk sac. Subsequently, the rate of development in the contaminated aquaria was more rapid than in the control. However, the mortality and number of deformities did not differ from those of the controls. At 10^{-8} and 10^{-6} Ci/liter, the rate of development was the same as that observed in the 10^{-10} Ci/liter experiment. Mortality and deformities were increased only at the higher level of radioactivity.

Studies by Kulikov *et al.* (1966) of the effects of ^{90}Sr-^{90}Y on the development of the eggs of the freshwater mollusc *Limnaea stagnalis* L. show that morphological abnormalities, delay in development, and mortality were significant only at concentrations greater than 10^{-4} Ci/liter.

Nelson (1968) studied the effects of radionuclides on Pacific oyster larvae, *Crassostrea gigas*. The larvae were reared for 48 hr following spawning in seawater containing either ^{65}Zn, ^{51}Cr, or ^{90}Sr-^{90}Y. The concentrations of each radionuclide range from 10^{-2} Ci/liter to 10^{-8} Ci/liter. Significant increases in abnormal larvae were detectable at the following minimal concentrations: ^{65}Zn (carrier free), 10^{-4} Ci/liter; ^{51}Cr, 10^{-4} Ci/liter; ^{90}Sr-^{90}Y, 10^{-3} Ci/liter.

The effects of tritiated seawater on the germination of the asexually produced spores of *Padina japonica* have been studied. A reduced percentage of germination was observed in those subjected to a concentration of tritium of 3×10^{-2} Ci/liter. This concentration of tritium also had a marked effect on the subsequent growth of the embryos, causing a decrease in the number and length of the rhizoids as well as in the number of cells in the multicellular filamentous thallus (Buggeln and Held, 1968).

All of these studies have assessed radiation effects in terms of observable gross effects. Ivanov (1967) has considered more sensitive parameters and reports on the effects of ^{90}Sr-^{90}Y in seawater on the mitotic activity and production of chromosome aberrations on the dividing cells of eggs of the Black Sea scorpionfish *Scorpaena porans*. As the concentration increased from 10^{-10} Ci/liter to 10^{-5} Ci/liter, the mitotic activity of the cells decreased. At the same time, the percentage of chromosome aberrations increased, with statistical significance when the concentration exceeded 10^{-9} Ci/liter. The types of aberrations were varied, with chromosomal and chromatid bridges and fragments observed most frequently. At the highest concentrations, abnormal mitoses were observed.

Experimental data concerning the radiation effects on developing embryos maintained under laboratory conditions in contaminated media are conflicting. In all these studies, the embryos were maintained under highly artificial conditions in the laboratory, including artificial fertilization. The developmental temperatures varied with species and experiments, ranging from 6°C to 24°C. In some experiments, antibiotics were used to reduce bacterial contamination. In addition, experimental incubation containers varied in capacity from 60-ml petri dishes to 40-liter plastic containers, with experimental lots ranging from 30 to thousands of individuals. The successful maintenance of marine eggs and embryos under controlled environmental conditions is yet to be developed for many marine organisms. In addition, more data are required on the effects of other environmental parameters before significant radiation effects can be demonstrated.

Of particular significance in the work from the Soviet Union is the unique concentration effect response (Polikarpov, 1966). An increase in concentration of ^{90}Sr-^{90}Y of six orders of magnitude (2×10^{-10} to 2×10^{-4} Ci/liter) no more than triples the abnormality production rate (Figure 3), and increases mortality only fivefold. If the radiation dose received by the developing eggs is proportional to the concentration and all other factors are equal, one would expect a more marked dose-effect response. The results are inconsistent with the linear hypothesis of dose response as well as with data from many radiobiological and

TABLE 3 Mortality, Growth, and Radionuclide Concentration in Chinook Salmon, *Oncorhynchus tshawytscha*, Reared under Various Reactor Effluent Conditions, December 1965-April 1966[a]

Treatment groups[b] (% effluent)	% Mortality (5 mo)	Mean Weight (g)	Concentration (pCi/g wet weight)		
			^{24}Na	^{51}Cr	^{65}Zn
0	18	0.70	77	19	6.8
2	10	0.82	750	38	20
4	13	1.05	1,390	53	36
6	13	1.20	2,210	65	45

[a] After Olson (1967).
[b] At least 1,000 fish in each group.

toxicological investigations. Although our knowledge of the dose-effect response at low levels of irradiation is limited, present knowledge suggests that perhaps factors other than radiation need to be considered and evaluated.

The biological effects of the effluent from production reactors at the Hanford plant have been monitored for more than 20 years by rearing salmonids in diluted effluent. The three main factors in Hanford reactor effluent considered to have a potential effect are the thermal increment, radioactivity, and chemical toxicity, the last from the hexavalent chromium used as a corrosion inhibitor (Olson and Nakatani, 1965; Nakatani and Foster, 1966; Olson, 1967).

Freshly fertilized eggs of salmonids have been incubated, and the fish reared in various concentrations of effluent until they reached migrant-sized fingerlings. The mortality, growth rate, and radionuclide concentration in chinook salmon reared under various effluent concentrations for 5 months during the 1965-1966 season are shown in Table 3. No significant lethality occurred in an effluent concentration of 6 percent, far above the existing levels in the river. The concentrations in the fish of the three gamma emitters, ^{24}Na, ^{51}Cr, and ^{65}Zn, are approximately proportional to the effluent concentration. The body burdens at these levels have produced no demonstrable damage in chinook salmon.

Internal Emitters

The effects on rainbow trout, *Salmo gairdneri*, of ingestion of some biologically important radionuclides have been studied. Yearling trout were force-fed radionuclides in capsule form over extended periods to determine what quantity of radionuclides must be ingested to produce radiation syndrome and damage. All of these experiments used very high levels of radionuclides, which would not normally be experienced in a contaminated aquatic environment, as the controlled disposal of radionuclides is limited to lower levels by man's use of the environment. However, the data from these ingestion experiments help to keep observations of the body burdens of radioactivity in field-contaminated fish in proper perspective with respect to potential radiation damage in fish in general.

In a series of feeding experiments, either ^{32}P, ^{90}Sr-^{90}Y, or ^{65}Zn was fed daily to rainbow trout at levels from 0.005 μCi/g/fish to high levels of about 10 μCi/g/fish (Watson *et al.*, 1959; Nakatani and Foster, 1963; Nakatani, 1966). Table 4 summarizes the results. The hematopoietic tissues of trout were found to be the most radiosensitive, as in mammals. Leukopenia was an early indicator of radiation damage following the ingestion of beta-emitting ^{32}P or ^{90}Sr-^{90}Y and gamma-emitting ^{65}Zn. Depression of growth also indicated radiation damage for the rapidly growing yearling trout. The radiation syndrome, including leukopenia, anorexia, loss of scales, lethargy, and growth depression, was very pronounced for fish fed ^{32}P at the higher levels and to a lesser extent for fish fed ^{90}Sr-^{90}Y. Because much of the energy from the gamma-emitting ^{65}Zn is not absorbed, trout were able to ingest without observable effect much higher levels of ^{65}Zn than of ^{32}P or ^{90}Sr-^{90}Y on a μCi/g/fish basis. In addition to the differences in the radiation characteristics of the isotopes, the rates at which the fish metabolize and the deposition sites for each radionuclide also differ, although all three are "bone-seekers."

Columbia River fish captured in the Hanford environs contain small amounts of virtually all of the radionuclides present in the water, but the only nuclides that accumulate in the fish flesh in significant amounts are ^{32}P and ^{65}Zn (Foster and Soldat, 1966). In 1965, the average concentration in muscle tissue of whitefish, *Prosopium williamsoni*, was 200 pCi/g and 27 pCi/g for ^{32}P and ^{65}Zn, respectively. Substantial seasonal variations may occur, with a tenfold difference for ^{32}P and a fourfold difference for ^{65}Zn. In terms of radiation damage to fish, however, the concentrations are far below those levels for which demonstrable damage to fish can be expected. Laboratory trout with a body burden of ^{65}Zn 10,000 times greater than that of river fish showed no detrimental effects. Similarly, trout with a body burden of ^{32}P 100 times greater than that of river fish showed no effect, although leukopenia occurred if the body burden was 1,000 times greater.

The determination of the actual dose absorbed by organisms and tissues is fundamental to the assessment of the potential effects of radiation in aquatic studies. Such determinations have been reported in only a few studies, however.

Although no dosimetric measurements were made for rainbow trout fed ^{65}Zn, Erickson (1966) calculated the

dose from data on body burdens of serially killed fish fed ^{65}Zn daily over a period of 16–18 weeks. He estimated a total absorbed dose of 25 R for trout fed 0.01 μCi/g of body weight and 1,200 R for trout fed 1.0 μCi/g. Watson *et al.* (1959) also calculated a dose to the bone, gut, and muscle of approximately 6,000 R, 1,600 R, and 800 R, respectively, in trout fed 0.06 μCi of ^{32}P/g daily for 17 weeks. At this level, the growth rate of the fish was reduced. No deaths were attributable to radiation damage, and there appeared to be an increase in growth rate after cessation of isotope feeding.

In some of the experiments involving chronic dosages to marine fish eggs, absorbed doses were measured and calculated (Table 5) (V. M. Brown, 1962; Brown and Templeton, 1964). Absorbed doses calculated for anchovy eggs from the basic data of Polikarpov and Ivanov (1962) indicate that at the concentration where abnormalities become significant (10^{-10} Ci/liter), the dose absorbed was 0.16 mR/day. The total dose accumulated by the eggs was the same, since the period from fertilization to hatching was 20–24 hr. The calculated dose rate from the ^{40}K in the seawater and in the egg was about 0.02 mR/day. However, consideration must also be given to the additional background dose within the laboratory, which would be about 0.3 mR/day, or approxi-

TABLE 4 Effect of Chronic Ingestion of ^{32}P, ^{90}Sr-^{90}Y, or ^{65}Zn on Yearling Rainbow Trout (*Salmo gairdneri*)[a]

Treatment (μCi/g fish/day)	Duration of Feeding (wk)	Growth Depression	Significant Mortality	Leukopenia	Gut Damage	Concentration at End of Feeding (μCi/g wet weight)	
						Bone	Muscle
^{32}P							
0.006	25	no	no	no	no	–	–
0.06	25	wk 17	no	4 mo	no	1.8	0.23
0.60	25	wk 11	yes	17 days	yes	–	–
^{90}Sr-^{90}Y							
0.005	21	no	no	no	no	2.1	0.0022
0.05	21	no	no	no	no	28	0.078
0.50	21	wk 12	wk 15	wk 15	yes	248	0.27
^{65}Zn							
0.01	17	no	no	no	no	–	–
0.10	17	no	no	no	no	–	–
1.0	17	no	no	no	no	4.0	0.35
10.0	10	no	no	wk 10	no	–	–

[a]After Watson *et al.* (1959); Nakatani and Foster (1963); Nakatani (1966).

TABLE 5 Calculated and Measured Radiation Dose Rates from Contaminated Media to Fish Eggs under Experimental Conditions[a]

Fish	Concentration Factor	Nuclide	Water Concentration (pCi/liter)	Range of Dose Rate to Egg (per day)	Dose Rate from ^{40}K in Water and Egg (mR/day)	Approximate Background Dose Rate in Laboratory (mR/day)
Salmon (*Salmo salar*)	30	^{90}Sr-^{90}Y	10^3	1–10 mR	0.017	0.3
Brown trout (*Salmo trutta*)	25	^{90}Sr-^{90}Y	10^5–10^{-7}	60 mR–3.0 R	0.017	0.3
Plaice (*Pleuronectes platessa*)	0.35 10.0	^{90}Sr ^{90}Y	10^2–10^8	<0.1 mR–13.0 R	0.02	0.3

[a]After Brown (1962) and Brown and Templeton (1964).

mately twice that calculated to be delivered to the egg from the ^{90}Sr-^{90}Y.

Calculations of the dose rates from radionuclides absorbed by or adsorbed on the organism in the laboratory and in the natural environment are difficult to compute accurately for several reasons. The radionuclide is not uniformly distributed throughout the organisms or even throughout the volume of a single organ or cell. One organ or tissue may contribute to the dose of an adjacent organ, the concentration may change with time, and the structures of organisms are of varying size and shape. A further complexity is the wide variation in the energy of the emissions. Calculations for the assessment of the dose to fish eggs arising from immersion in contaminated seawater have been developed by Fedorov (1965) and Adams (1968).

A major advance in the determination of dose is the development of thermoluminescent dosimeters (TLD's). These dosimeters are commercially available in a variety of forms, most commonly as LiF powder in sachets or as LiF extrusions. The lower limits of detection in routine use are of the order of 20 mR, with an error of about 10 percent, and the techniques are rapidly being improved. The small size of the dosimeters allows them to be conveniently emplaced on or in living organisms to provide the best practical means of direct measurement of the radiation dose received by the organisms in their natural habitat. These dosimeters can also be used to measure the radiation dose in sediments directly, and they can be used in water with inexpensive buoyline-anchor support systems.

The performance of TLD's is very good. They are little affected by wide variations in environmental conditions and can therefore be left unattended for long periods. They are nearly energy-independent over a wide range of energies and can be reused.

The principal limitations of TLD's, however, are the same as those of other dosimeters that measure accumulated dose—only the total dose can be determined without knowledge of the rate at any time during the exposure period or of the radionuclides contributing to the dose.

Thermoluminescent dosimeters have been successfully used to measure the dose to fish in the Windscale area (Ministry of Agriculture, Fisheries and Food, 1967, 1968) and to aquatic organisms of the Columbia River in the vicinity of the Hanford Works (Watson, personal communication). Tests of TLD's in the tropical marine environment using a ^{60}Co source indicate that 0.7-MeV gamma emitters at concentrations of approximately 10^{-3} μCi/ml in seawater could be detected after 48-hr exposures (Baltzo and Held, 1968). It should be expected that the results of more such studies, relating concentration to radiation dose in the aquatic environment, will appear in the literature.

With the wider application of this technique, the results of experimental studies and field studies, and, it is hoped, of the predictions of effect, will become more meaningful in the future.

INFLUENCE OF ENVIRONMENTAL FACTORS ON RADIATION EFFECTS

The potential ecological interactions and the assessment of the radiosensitivity of marine ecosystems have been discussed in a previous chapter. The influence of two major environmental factors—temperature and salinity—on radiation effects on marine organisms will be briefly reviewed here. The effect of oxygen on radiosensitivity of organisms has been studied extensively (Bacq and Alexander, 1961) and will not be reviewed here. The literature shows little experimental work on the interactions of temperature and salinity with radiation effects, but research into this subject becomes increasingly important with greater use of nuclear energy to meet man's needs for power and for fresh water from the sea. Nuclear power and desalination plants imply not only some release of radioactive materials to the sea but also release of heat and highly saline water.

As the metabolism of poikilotherms is dependent on temperature, it is easy to understand the greater radiosensitivity observed following elevation of the temperature during or after radiation exposure. After exposure to 28,000 R, fecundity of a freshwater snail, *Physa acuta*, was reduced to 30 percent of that of the control snails in 20°C water and to 12 percent in 30°C water (Ravera, 1966). The hatchability of *Artemia* eggs exposed to irradiation by x rays was significantly reduced in warm water, but when cooler water was used, no significant reduction occurred (Iwasaki, 1964; Cervini and Giavelli, 1965). Goldfish, *Carassius auratus*, exposed to 8,000 R all died within 10 days when maintained at 22°C, but similarly irradiated fish survived for more than 100 days at 4°C (Hyodo, 1965). Gros *et al.* (1958) postulated that low temperatures (7°C) would protect the crucian carp, *Carassius carassius*, against radiation damage and that radiation, on the other hand, would protect the fish against cold by lowering the thermal tolerance.

White *et al.* (1967) reported on the combined effects of ionizing radiation, salinity, and temperature on the estuarine fish *Fundulus heteroclitus*. In a factorial experiment, fish were subjected to four levels of acute radiation (500, 1,000, 2,000, and 2,500 rads), three levels of salinity (5, 15, and 25°/$_{oo}$), and four levels of temperature (12°, 17°, 22°, and 27°C). They found that different combinations of levels of temperature and salinity yield different LD_{50} values. The estimated LD_{50} values for different experimental conditions ranged from 300–350 rads to more than 2,500 rads. The significance of the experiments is the demonstration of the importance of the environmental factors on the radiosensitivity of aquatic organisms. In other words, the effects of radiation on aquatic organisms can be evaluated only along with the effects of other major environmental factors.

The investigations by Egami and Etoh (1966) and Etoh and Egami (1967) of the effect of temperature on the rate of damage accumulation and recovery in the fish *Oryzias latipes* showed that the processes of recovery from damage

induced by external irradiations (x rays or ^{60}Co γ rays) are active to some extent at 23°C but remain almost undeveloped at 11°C. For poikilotherms, it seems likely that low temperature not only delays the development of radiation-induced damage but also decreases the rate of recovery (Egami *et al.*, 1967).

Fish with body burdens of ^{90}Sr and ^{131}I were subjected to thermal shock of 13°C from an acclimation temperature of 25°C. These body burdens did not impair their ability to withstand lethal temperatures. Indeed, there was an indication that their survival time was increased. The approximate beta dose to the bone and thyroid tissues was calculated for the fish containing maximum concentrations of ^{90}Sr and ^{131}I. These were found to be at least 10^4 rads to bone tissue and 10^5 rads to thyroid tissues (Ophel and Judd, 1966). Blaylock and Mitchell (1969) determined that temperature was an important factor in the $LD_{50/30}$ for *Gambusia affinis affinis*, since a difference of only 5°C resulted in a significantly different $LD_{50/30}$.

Few experimental data are available to explain how temperature, salinity, and other physicochemical environmental factors interact to affect the radiosensitivity of different aquatic organisms. It is recognized, however, that all organisms in nature have always been subject to different environmental stresses, including radiation, in varying degrees. The possibilities for damage to aquatic organisms will most probably arise through the combination and interaction of different environmental factors, since survival is rarely dependent on a single environmental factor.

BEHAVIOR AND METABOLIC STIMULATION

Scattered reports describe the use of behavioral criteria to determine the effects of radiation on aquatic organisms. These studies suggest that aquatic organisms apparently detect ionizing radiation, although the receptors have not been identified. In experiments with fish, particularly for high levels of external radiation, it has not been established whether the fish are responding directly to a radiation source or indirectly to induced products of water hydrolysis. O'Brien and Fujihara (1963) observed that larvae of the freshwater cichlid fish *Aequidens portalegrensis* became hyperactive during x-ray irradiation. Doses of 50 R stimulated activity, and larvae remained active for 90 minutes after irradiation. These larvae are not normally free-swimming but remain in clumps on the bottom. O'Brien and Fujihara also observed the influence of a ^{137}Cs source in the water, emitting less than 5 rads per hour at a distance of 1 in., on the behavior of 2-day-old cichlid larvae. The control larvae were scattered randomly around a sham source, but the experimental larvae avoided a ^{137}Cs source. Pravdina (1965) reports that carp avoided x-ray sources providing a dose rate of 2.5 rad/min.

Hyperactivity in fish has also been reported by Scarborough and Addison (1962). They observed definite periods of hyperactivity in the golden shiner, *Notemigonus crysoleucas*, during irradiation at doses of 7,200–18,000-R x rays given for periods of 6–12 min. Tsypin and Kholodov (1964) used γ rays from ^{60}Co at a dose rate of 0.1–0.5 rad/sec for a duration of 5–10 sec as a conditioned stimulus, and they used electric shock as the unconditioned stimulus on fish. Four of thirteen trials resulted in a motor response to the γ rays alone. A modification of the locomotor orientation of the turbellarian *Dugesia dorotocephala* has been reported under low-level chronic irradiation in the range 26 μR/hr to 240 μR/hr (Brown, 1962; Brown and Park, 1964). Field *et al.* (1964) compared the swimming activity of rainbow trout tagged with 16-μCi ^{60}Co wire tags and with nonradioactive tags. Tests made 16 hours after tagging showed a significantly higher swimming activity in ^{60}Co-tagged fish, which persisted for at least 26 hours.

Apparent stimulation of growth by radiation in aquatic organisms is reported for periphyton, certain marine invertebrates, young blue crabs, and rainbow trout. The growth of periphyton during 5 months in aquaria containing mixed fission products (3×10^{-6} to 6×10^{-7} Ci/liter) resulted in a greater biomass and greater species diversity than in control aquaria (Timofeyeva-Resovskaya, 1958). Glass plates with ^{90}Y on the surface at a concentration of 3.3 Ci/cm^2, with a calculated surface dose of 40 rads/day, exposed in the sea, enhanced the development of barnacles, bryozoans, calcareous worms, and small mussels (Dolgopolskaya *et al.*, 1959).

Engel (1967) reports that exposure of young blue crabs, *Callinectes sapidus*, to ^{60}Co at 3.2 rads/hr resulted in significantly more rapid growth than that of crabs irradiated at 7.3 and 29.0 rads/hr or of control crabs. Since molting is associated with growth, the percentage increase in width of molted carapaces was monitored; it was found to be related to the radiation dose rate. Crabs receiving 29.0 rads/hr, the highest dose rate, had the smallest percentage of increase in carapace width, and those that received 3.2 rads/hr had the largest increase in width, compared with the control crabs.

Metabolic activity of salmon (*Salmo salar*) eggs and fry was increased by the presence of ^{137}Cs at a concentration of 1 μCi/liter. Oxygen consumption increased gradually throughout development and was always higher in the ^{137}Cs-treated eggs than in the controls. Upon hatching of the eggs, the oxygen consumption increased by 50 percent in the controls and 200 percent in the treated aquaria (Neustroev and Podymakhin, 1966b). Although insufficient replicate groups were maintained to provide a rigorous statistical test, rainbow trout fed 0.01, 0.1, and 1.0 μCi ^{65}Zn/g of fish daily for 17 weeks showed more rapid growth than the control group (Nakatani, 1966).

Other examples of possible stimulation of organisms by radiation under laboratory conditions can be found; however, little or no knowledge exists about either the mechanisms involved or the significance of radiation as a possible stimulus to individuals, populations, or ecosystems.

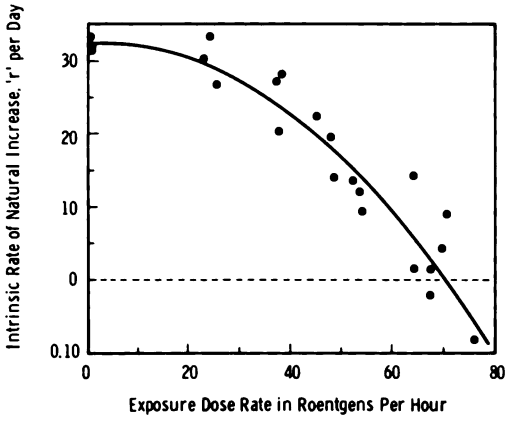

FIGURE 4 The effects of continuous gamma radiation on the intrinsic rate of natural increase of *Daphnia pulex*. (Reprinted with permission from Marshall, 1962.)

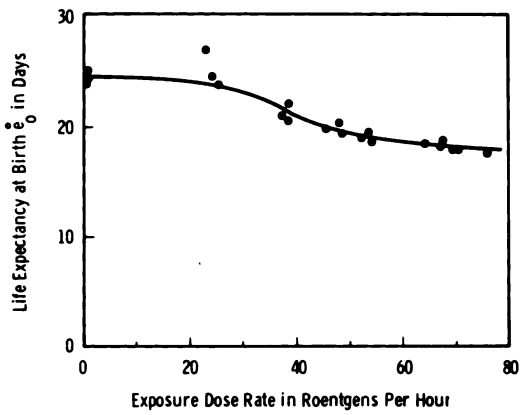

FIGURE 5 The effects of continuous gamma radiation on the life expectancy at birth, $\mathring{e}_0$, or average life-span, of *Daphnia pulex*. (Reprinted with permission from Marshall, 1962.)

RADIATION EFFECTS ON POPULATIONS

Ultimately, we are concerned with radiation effects on populations and ecosystems in the marine environment rather than with the demise of individuals. Marshall's (1962) work with *Daphnia* is indicative of a line study that is particularly significant. He investigated the effects of gamma radiation on the intrinsic rate of natural increase, *r*. The dose rates used ranged from 25 to 75 R/hr, and the cultures were exposed for about 19 hr/day.

Reproduction was entirely parthenogenetic. There was a continuous decrease of *r* as a nonlinear function of the dose rate (Figure 4). The decrease was almost entirely caused by a falling birth rate, which in turn resulted from direct effects of radiation on the ovaries. Average life-span was not greatly shortened, even at the highest dose (Figure 5). Interestingly, growth in length of individuals increased with increasing dose rate, which was interpreted as due to the utilization for growth of energy ordinarily spent in egg production.

Grosch (1962, 1966) followed the reproductive capacity of mass cultures of *Artemia* for 8 years. Cultures were repeatedly contaminated with sublethal amounts of ^{65}Zn or ^{32}P (~7-30 μCi/liter). The following are some of Grosch's conclusions:

> Although the number of adults seen in mass cultures may be equivalent, subcultures of control and experimental strains react differently to radioisotope additions. Strains descended from ancestors exposed to ^{32}P do not necessarily survive a second dose even though total dosage does not exceed the extinction dose given as a single addition. A period of recovery involving generations must intervene. Most notable is the consistent demonstration that the number of adults can be identical in different mass cultures, but that the reproductive potential of populations with different ancestral histories differs considerably. On the basis of pair mating tests, maintenance of mass cultures at an observed level of 300 adults per three liters requires only 0.2% of the reproductive potential of controls. Cultures of experimental origin utilize 1% or more of their potential to maintain the same total.

Grosch has a detailed discussion in which he compares his conclusions with those derived from *Drosophila* populations (Wallace, 1956) and *Tribolium* (Crenshaw, 1965), which showed increased genetic fitness following the irradiation of inbred strains.

ENVIRONMENTAL STUDIES

Pacific Proving Grounds

The first large-scale introduction of man-made radionuclides into a marine environment was at Bikini Atoll in 1946. Two 20-kiloton devices were detonated, the first an air burst and the second an underwater detonation in the 250-mi^2 lagoon, which has maximum depth of about 60 m. In succeeding years, through and including 1958, Bikini and Eniwetok became the Pacific Proving Ground. During that time, nuclear and thermonuclear devices with a total yield of many megatons were detonated at the atolls. Certainly, these atolls represent the most radioactively contaminated marine environment in the world, as far as is known from public announcements. And yet today, more than 23 years since the initial contamination of the atolls, a statement by Schultz in a 1947 report on the observed biological effects of the nuclear weapon test "Operation Crossroads" still holds true:

> Undoubtedly, countless animal individuals have perished at Bikini because of the atomic bomb experiments and still others may perish. But, this destruction of life in a large

atoll like Bikini amounts to only an extremely small percentage of the total animal life. The overall picture of life on the reefs has changed little because beneath this surface layer, and from extensive adjoining unaffected areas, individuals have come forth to repopulate and occupy the reefs. The pressure of population from all sides into the damaged areas is very great and soon replaces the losses. Thus, nature begins the repopulation cycle, and, if given sufficient time, the wounded reefs will be cleansed of their contamination, biological equilibrium will be reached; and life will establish itself as in past millenniums–similar to that before man released the greatest destructive force in his history.

It is inconceivable that there were no radiation effects at the test sites. Evidently, where the prompt radiation at the moment of detonation was sufficiently intense to produce immediate visible effects, the concomitant effects of blast and heat virtually eliminated the populations. Furthermore, those individuals suffering sufficient injury from the residual radiation to be readily recognized are soon eliminated. In other words, gross radiation injury in marine organisms has not been seen at Bikini and Eniwetok because seriously injured individuals do not survive the natural rigors of the environment, and the more subtle injuries are exceedingly difficult to detect.

Bikini and Eniwetok were intensively studied before and following the test series (Revelle, 1954; Hines, 1962), including extensive studies of fish (Schultz *et al.*, 1953, 1960; Hiatt and Strassburg, 1960; Welander, 1957; Welander *et al.*, 1967), corals (Wells, 1954), and algae (Taylor, 1950; Dawson, 1957), and yet in none of the reports on marine organisms is there reference to anomalous individuals.

However, Gorbman and James (1963) studied the thyroid histologically and observed thyroid tissue damage in fish collected at Eniwetok Atoll 30 days to 8 months following nuclear detonations. Although no radioiodine was detectable in the fish at the time of examination, it seems clear from indirect evidence that the observed anomalies were caused by radioiodine. Blinks (1952) examined physiological functions of sessile algae at Bikini a year after the first atomic tests; at that time, the dose rate was estimated to average 20 to 30 mR/day. He concluded that there was "no noticeable alteration of many normal somatic functions."

The land plants and animals were subjected to greater intensities of radiation both from external sources and from internally deposited radionuclides, but even here, lasting effects of radiation on populations or on the ecosystem are not apparent.

Jackson (1967) has reconstructed the story of the survival of rat populations at Eniwetok Atoll. Engebi Islet, with an area of 260 acres, was subjected to radiation in 1948, 1952, 1954, 1956, and 1958. The accumulated dose a year after contamination was 44 R (1948), 11,000 R (1952), and 118 R (1954). Prior to the nuclear tests, probably only the Polynesian rat (*Rattus exulans*) was present at Engebi, but early in the testing period the roof rat (*R. rattus*) was unintentionally introduced. The former makes a nest under surface vegetation and debris, while the roof rat often burrows. Rats were killed following the detonations in 1948, 1952, and 1954, but a sufficient number of individuals survived to re-establish the population. In summary, Jackson states:

> The Eniwetok story is made difficult by taxonomic confusion and lack of specimens. At best, a hypothetical reconstruction can be attempted. Early in the test program at Engebi Islet, the Polynesian rat (*Rattus exulans*) was exterminated, probably by heavy surface radiation. Prior to the detonation of a thermonuclear detonation in 1952 [14 megatons detonated approximately 3 mi from Engebi], the roof rat (*R. rattus*) had become established on Engebi; and a nucleus survived the heavy initial radiation by being deep in burrows. Calculations show that repopulation by 1954 and 1955 was theoretically possible. The decline of the Engebi rat population from its high density in the mid-fifties was a result, probably, of a change in the carrying capacity of the environment.

Cole (1951) reported that the insects of Bikini were studied for structural anomalies in 1947 and that none were found. He further stated that "continuing studies of *Drosophila* cultures, taken in the living form at Bikini Island, have not thus far [1947-1951] revealed genetic abnormalities in excess of normal variability." However, more detailed studies following further contamination did reveal probable genetic effects in *Drosophila*. Stone and his co-workers (Stone *et al.*, 1957; Stone and Wilson, 1958, 1959; Stone *et al.*, 1962) made extensive genetic studies of wild populations of *Drosophila* subjected to fallout in 1954 at Bikini, Rongelap, and Rongerik Atolls, comparing them with populations of uncontaminated islands. The estimated gamma dose to the Bikini *Drosophila* population was 3,000-8,000 R, but the absorbed dose is not known. The conclusion was reached that the genetic load of detrimental and lethal factors of the irradiated *Drosophila* populations had been increased but was returning to a normal range. These tests were possible only because of the extensive and sophisticated genetic techniques for *Drosophila*. Similar established techniques are not now available for the marine organisms; hence, we can only speculate that they, too, might have had an increased deleterious genetic load but have recovered or are continuing to recover.

Rongelap Atoll was heavily contaminated with radioactive fallout in 1954 (Glasstone, 1962; Dunning, 1957). From observations in 1956, Fosberg (1959a and b) described the condition of plants at islets where the integrated dose was approximately 3,360 R. He suggested that the situation observed might be primarily due to the radiation. Held (1963a), following further studies at Rongelap, suggested that other environmental factors might be more important than radiation in this case. The question is not resolved, nor is it likely to be without controlled experiments with the species involved under varying conditions.

In a more intensive study, Palumbo (1962) documented the recovery of land plants at a site 2½ miles from a multimegaton detonation at Eniwetok. The vegetation was destroyed by heat and blast, and the estimated external dose to the vegetation was 400 R in 200 days. Palumbo noted that regrowth of the vegetation occurred in 6 months and described two minor anomalies. Palumbo discusses many of the factors influencing plant growth in the island environment and reviews the anomalies in plants reported by others at the test site and in other areas of the Central Pacific. He points out that similar anomalies have been observed in areas of heavy fallout and in uncontaminated areas. Again, there can be no doubt that there was damage and even the destruction of individual plants by radiation; but the specific examples of damage or killing caused by radiation or by other factors, or in combination with other factors, cannot be sorted out. The important point is that although individuals may be debilitated or destroyed, the ecosystem recovers.

The land-dwelling hermit crab, *Coenobita* sp., and coconut crab, *Birgus latro*, are subject to higher levels of chronic radiation from internally deposited radionuclides than any other organism studied at the atolls. The levels of ^{90}Sr and ^{137}Cs were found to remain virtually constant at 4,500 pCi of ^{90}Sr per gram of skeleton and 450 pCi of ^{137}Cs per gram of muscle in *Coenobita* sp. at Eniwetok over a period of two years (Held, 1960).

Parallel studies of *Birgus* sp. at Rongelap Atoll showed that the crabs contained more than 700 pCi ^{90}Sr per gram of skeleton and 100 pCi of ^{137}Cs per gram of muscle over a period of 10 years, 1954–1963. No gross anomalies were noted among these crabs, and no obvious population changes were noted during this fallout period; however, population studies as such were not made.

Observations at Bikini Atoll in 1964 (Welander *et al.*, 1967) showed that vegetation nearly covered the islet in a mass that was impenetrable without cutting. The vegetation, exclusive of roots, taken from an area 10 m in diameter in a *Scaevola* sp. community in 1967 yielded 108 kg/hectare (wet weight), and no gross anomalies were seen on the islet (Held, 1967, unpublished field notes). The abundance and size of fish and of spiny lobsters and coconut crabs at Bikini Atoll appear to be greater than ever, which does not, of course, reflect a beneficial effect of radiation but presumably results from the absence of predation by man.

Irish Sea

Apart from the Pacific Proving Grounds, the Irish Sea coastal area adjacent to the Windscale reprocessing plant of the United Kingdom Atomic Energy Authority probably represents the most important area known with respect to the degree of contamination in the marine environment (Mauchline and Templeton, 1964).

Studies of the relationships between radionuclides and man have been of prime importance; some studies, though limited, have considered effects of radiation in the environment. Morgan (1960) reported that effects of radiation had been sought in plaice (*Pleuronectes platessa*) caught in the region of the Windscale discharge, but none has been established so far. The area, which had been carefully surveyed before and after discharges began, showed that there were no changes in bottom organisms that could be ascribed to the effects of the discharge.

Calculations (Dunster *et al.*, 1964) of the external radiation dose rate to benthic organisms that would result if the seabed were at the derived working limit of 0.1 μCi/g total beta activity are about 45 mR/hr, or 1 R/day, mainly of beta radiation.

In a series of experiments concerned with the uptake by plaice eggs of six radionuclides, ^{85}Sr, ^{137}Cs, ^{144}Ce, ^{103}Ru, ^{88}Y, and $^{95}Zr/^{95}Nb$, data were obtained on the rates of accumulation and concentration factors (Woodhead, 1970). The concentration of these radionuclides in the egg can now be calculated for any concentration of the radionuclides in seawater to which they may be exposed. Tentative conclusions have been drawn that the dose rate to the eggs of plaice from the contaminant activity in the spawning areas off St. Bees Head, near Windscale, was 9.1×10^{-2} μrad/hr compared with 7.0×10^{-1} μrad/hr from the natural ^{40}K in the seawater.

Studies have also been made of the radiation dose received by fish in the Windscale discharge area (Ministry of Agriculture, Fisheries and Food, 1967). The plaice, *Pleuronectes platessa*, is a seabed resident and is known to spend its early life, up to the beginning of its third year, inshore. Calculations were made of the potential dose that a fish could receive on the basis of data from measurements of seawater, seabed, and the fish themselves over the previous 5 years at a point 2 miles south of the outfall and 1½ miles offshore from the pipeline. The radionuclides ^{137}Cs, ^{106}Ru, $^{95}Zr/^{95}Nb$, ^{144}Ce, and ^{90}Sr were considered, since these represent the major contributions to dose in terms of the various known factors of concentration in the major segments of the environment. In the calculations, allowance was made for fish movement and type of bottom, and these indicated that an annual dose of 7.3 rads could be accumulated, with most of the dose contributed by the seabed (Woodhead, 1968 personal communication). The seawater and internal radionuclides would only contribute about 1 percent of the total expected dose (Table 6). The annual dose might increase to 40–50 rads if the fish spent the whole year close to the pipeline outlet on a silty bottom.

This calculation suggested that the use of thermoluminescent dosimetry was warranted, and 2,500 marked plaice were released in the vicinity of the outfall in 1967 (Ministry

TABLE 6 Estimates of Dose Rates to Plaice from Fission Products at Windscale[a]

Source	Type of Radiation	mR/week
Seawater	γ	0.01
	β	0.01
Seabed	γ	48.8
	β	91.4
Radionuclides in muscle	γ	0.013
	β	0.057
		140.3[b]

[a] Data from Woodhead (1968, personal communication).
[b] Or 7.3 R/year.

of Agriculture, Fisheries and Food, 1968). Each fish carried two lithium fluoride dosimeters, incorporated in a Petersen fish tag, one to measure the accumulated dose on the upper surface and one to measure the dose on the under surface. In the first few months, over 243 marked fish were recovered, and preliminary data indicated an integrated exposure of 4.5 R for the bottom dosimeter. This was equivalent to a dose of 10 R per year. The ratio of top to bottom dose was 0.73. The differential response of dosimeters placed on the top and the underside of the fish indicated the expected response to beta radiation. The agreement between calculated and measured dose is extremely good.

Gamma dose rates measured 1 m above the Ravenglass mud flats, near Windscale, from January 1965 to June 1967, averaged about 140 μR/hr, compared with 10-15 μR/hr background. Over sandy beaches, dose rates ranged from 11-16 μR/hr, compared with a background rate of 8-12 μR/hr. The mean dose rate over mud, from $^{106}Ru/^{106}Rh$, $^{144}Ce/^{144}Pr$, and $^{95}Zr/^{95}Nb$, has been calculated as about 1 mR/hr, or 8 R/yr (Jeffries, 1968).

Thermoluminescent dosimetry measurements in 1965, using LiF powder in sachets implanted in the mud at a point where $^{106}Ru/^{106}Rh + ^{144}Ce/^{144}Pr$ levels were about half those reported by Jeffries, gave values of 100 mR/wk in the top 1 in. of mud, 50 mR/wk at a depth of 3 in., and <20 mR/wk at depths to 17 in. Organisms in the top 3 in. might then be exposed to a radiation dose of 4.0 R/yr, comparable to that received by fish near the outlet (Templeton, 1968, personal communication).

Oak Ridge

Very few studies have been made of natural populations exposed to chronic radiation higher than background. The salivary chromosomes of the larvae of *Chironomus tentans*, which inhabit the contaminated bottom sediments of White Oak Creek and White Oak Lake at Oak Ridge National Laboratory, were analyzed for 5 years for chromosomal aberrations (Blaylock, 1966). Calculations and measurements of the adsorbed dose for the larvae living in the sediments gave values of 230-240 R/yr, or 1,000 times background for that area. More than 130 generations had been exposed to this or greater dose rates over the previous 22 years. The conclusion was that the ionizing radiation from the contaminated environment was increasing the frequency of new chromosomal aberrations in the irradiated population, but that the new aberrations were eliminated by natural selection. Also, the present level of chronic irradiation has not affected the frequency of the endemic inversions.

Blaylock (1969) also studied the fecundity of a natural population of fish, *Gambusia affinis affinis*, that had been exposed to chronic irradiation in White Oak Creek for many generations, compared with a control population. The calculated dose rate from the bottom sediments was 10.9 rads/day. A significantly larger brood size occurred in the irradiated than in the nonirradiated population, although significantly more dead embryos and abnormalities were observed in the irradiated broods. These results suggest that an increased fecundity is a means by which a natural population having a relatively short life cycle and producing a large number of progeny can adjust rapidly to an increased environmental stress caused by radiation.

We return then to a concept that has recurred in this report: Man, as an individual, is the critical biological target in predicting the consequences of introducing radioactive materials into marine environments. If the radionuclides are present in concentrations acceptable for man, the individual, then it is difficult to conceive that there will be more than subtle effects on ecosystems—perturbations that would probably be indistinguishable from those due to causes other than radiation. On the other hand, it is not only conceivable, but probable, that with increasing uses of atomic energy, accidents will occur that will result in damaging concentrations of radionuclides. In preparation for these contingencies, there is a pressing need to increase the sensitivity of methods for studying the response to radiation of populations and ecosystems in the marine environment.

EFFECTS ON RESOURCES

The extrapolation of the results of laboratory experiments into the practical terms of their effects on marine resources must be made with care since, without evaluation of the natural variations related to changes in fecundity, mortality, and recruitment, quite erroneous conclusions can be reached. Polikarpov (1966) suggests "inhibition and degeneration of a number of food fishes and other radio-sensitive organisms" and "rapid development and multiplication of

bacteria, microphytes and radio-resistant forms of invertebrates" as a result of radiation modification of the marine environment.

Zaytsev and Polikarpov (1964) and Polikarpov (1966), based on their previously discussed laboratory data, have calculated the time needed to halve the stocks of various species of Black Sea fishes in relation to the proportion of the eggs damaged by radiation. These workers conclude that there is reason to assume that the fisheries will be adversely affected, and may, in many instances, cease to function if fish stocks are reduced to one half or less. They consider that from the commercial standpoint, radiation damage must not affect more than 10 percent of the eggs, though even at this level, catches will be perceptibly reduced.

Garrod (1966) points out the fallacy of this argument. For marine mammals or for certain fish species with low fecundity, a 50 percent mortality of the young or of the eggs will be reflected as a 50 percent reduction in recruits to the stocks, though this does not generally occur in the marine environment. In the general case of organisms with high fecundity, reduction in the spawning stock does not necessarily result in a decrease in the number of recruits. The capacity of the environment to support young fish is limited, and this capacity can be satisfied by the eggs from a small number of adults, the superfluous eggs and larvae being reduced by density-dependent mortality.

The magnitude of the mortality is also pertinent in the evaluation of the effects of radiation on marine resources. Garrod indicates that for stocks of arctic cod with high fecundity, survival is 1 in 10^4 and can be as low as 1 in 10^5. The addition of 50 percent mortality in the egg stage due to the effects of radioactive contamination of the environment would not noticeably alter the existing mortality rate of 99.99 percent. This would also be true of all highly fecund species.

While it may be true that the fecundity of both the individual and the population could be reduced by radiation effects, it is extremely doubtful that these would influence stock size beyond the normal range related to environmental changes, except in very heavily exploited stocks. It is certain, however, that with the techniques of assessment now available, it would not be possible to obtain an unbiased measure of the effect attributable to radiation alone.

RESEARCH NEEDS

- Further studies are required concerning the concentrations of naturally occurring radionuclides and natural background radiation doses in the environment as a baseline for studies of the effects of radiation.
- Time curves for acute exposures of lethal doses should be extended, with particular attention to exposure as a function of age.
- Because of the conflicting nature of the present experimental data on the effects of low-level chronic irradiation on developing embryos, more studies should be established under rigorous, controlled experimental conditions. The effects of other environmental "stress" factors, such as salinity, temperature, oxygen, and pollutants, must be studied and expanded to include the interaction of these factors with radiation effects.
- More sensitive parameters of radiation effects on individuals, populations, and communities should be developed. Long-term studies, in which factors such as rates of growth, morphological abnormalities, onset of maturity, and reproductive capacity are considered, for example, should be emphasized. Chromosomal studies under experimental and field conditions should be extended, and the somatic and genetic consequences of such changes on populations and ecosystems should be evaluated.
- Measurements of absorbed dose using microdosimeters should become standard practice in experimental work. In areas subject to radioactive contamination, studies should be initiated to determine the radiation regime in the environment. The data obtained should be closely correlated with measurements of concentration of radionuclides in order that the historical radiation regime can be determined.

SUMMARY

Radiation is not a recent introduction to the marine environment, since low levels from environmental and cosmic sources have been present throughout geological time. Lethal amounts of acute radiation differ widely among marine organisms and are related to variations such as species, age, physiological status, and body size. These variations are further complicated by the interaction of environmental factors such as temperature and salinity. Exclusive of the eggs of fish and larvae of invertebrates and fish, most marine organisms for which data exist are relatively radioresistant.

Limited studies on the effects of chronic exposure have been conducted. These have been limited to selected developmental stages and indicate that, with the possible exception of some Russian data, the dose necessary to evoke an unequivocally detectable biological response is considerably above that of concentrations of radionuclides in the environment as a result of controlled waste disposal operations.

Studies on the genetic consequences of radiation exposure to population indicate that, despite larger numbers of mutations, increased utilization of reproductive capacity maintains a population at preradiation density.

Field studies on the effects of radiation indicate that our best technologies and methods cannot demonstrate effects on marine ecosystems, at prevailing dose rates, that are clearly and uniquely attributable to ionizing radiation.

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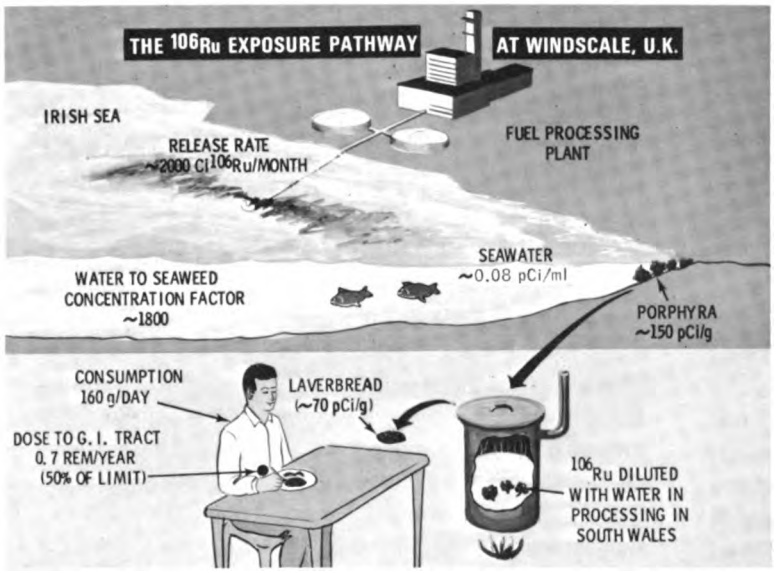

FIGURE 2 The critical exposure pathway associated with the U.K. atomic energy plant at Windscale is the release of ^{106}Ru to the Irish Sea, its accumulation by seaweed, and the consumption of the seaweed by man.

this period (Mitchell, 1967b) were less than 150 μR/hr (i.e., less than 2.5 percent of the acceptable dose rate when handling times for commercial fishing gear are considered).

Hanford Project The Hanford Project (now known as Richland Operations) discharges substantial quantities of radionuclides into the Columbia River. Some of these radionuclides reach the mouth of the river and enter the Pacific Ocean (Soldat and Essig, 1966). Of those radionuclides that reach the sea, only ^{65}Zn and ^{32}P warrant consideration as sources of exposure to humans in nearby communities. Oysters grown in a bay several miles north of the river mouth contain higher concentrations of these radionuclides than other common seafoods. Estimated human dose rates from an assumed consumption rate of 230 g of oysters per week are 5 mrem per year to the intestinal tract and 3 mrem per year to the whole body (Soldat and Essig, 1966). These represent 0.3 percent and 0.6 percent, respectively, of the acceptable dose rate to individual members of the public.

Other marine fish and shellfish in the vicinity also contain measurable concentrations of ^{65}Zn, but human exposure from these sources is much less than from oyster consumption.

Within the Columbia River, near the point of discharge of the effluent from the reactors, the uptake of ^{32}P by fish is the critical pathway. This is illustrated in Figure 3.

OTHER INSTALLATIONS

Radionuclides are discharged into the sea at a number of other research centers, including Trombay, India (Pillai and Ganguly, 1961); Studsvik, Sweden (Agnedal and Bergström, 1966); and Petten, Netherlands (Van Dam and Davids, 1966). Discharges at all these locations have been small in amount, and the critical pathways for human radiation exposure have been evaluated only as a preoperational exercise.

At Trombay, the critical radionuclide is ^{32}P, and the critical pathway is expected to be the eating of marine fish. At Studsvik, the composition of the discharged effluent is uncertain. Present indications are that external exposure from ^{60}Co on fishing gear will be the critical pathway for neutron activation products, and internal exposure from ^{106}Ru in fish will be the critical pathway for fission products. At Petten, the critical pathways are assumed to be fish and shellfish consumption. No critical radionuclides have been identified.

NUCLEAR POWER REACTORS

Bradwell The nuclear power station at Bradwell, England, discharges radionuclides into a river estuary. Here, the critical pathway is the consumption of contaminated oyster flesh, and the critical radionuclide in the effluent is ^{65}Zn (Preston, 1967) (Figure 4). Preoperational and follow-up assessments have been made (Table 3). The radiation dose received by the small critical population group is 0.17 percent of the acceptable dose rate for the total body and 0.08 percent of that for the intestinal tract (Preston, 1968).

Other U.K. Power Reactors At five other power reactor sites in the United Kingdom that began operation in 1968, preoperational assessments had been made of the critical pathways and critical radionuclides (Table 4). Actual discharges at Berkeley and Hinkley have been so low in relation to the maximum rate permissible for the site that only very small concentrations of radionuclides have been detected in the environment. At Berkeley, ^{137}Cs has been measured ex-

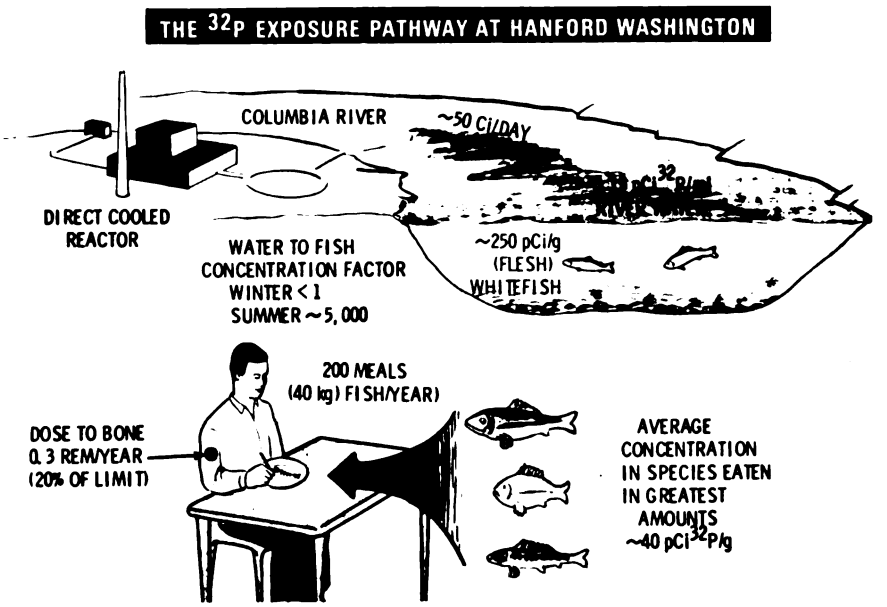

FIGURE 3 The critical exposure pathway associated with the Hanford plutonium-producing reactors is the release of ^{32}P to the Columbia River, its uptake by fish, and the consumption of the fish by man.

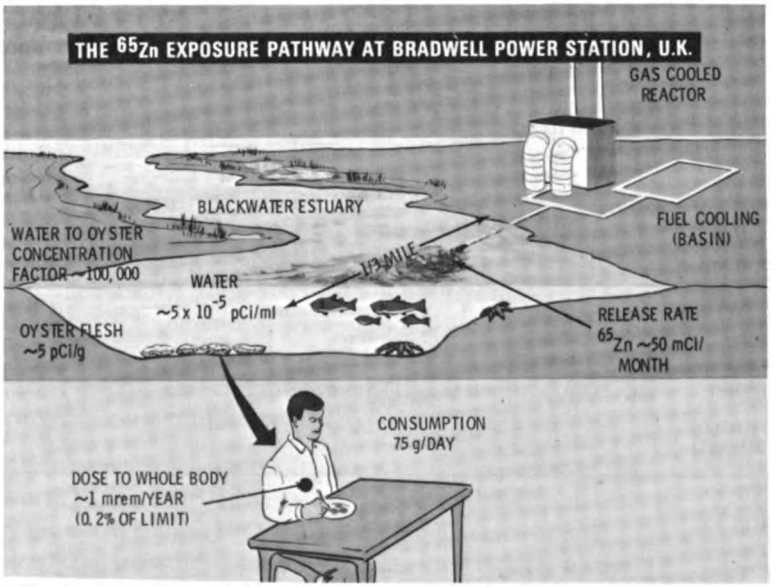

FIGURE 4 The critical exposure pathway associated with the nuclear power station at Bradwell, U.K., is the release of ^{65}Zn to the Blackwater Estuary, its uptake by oysters, and the consumption of the oysters by man.

tensively in silt (1–2 pCi/g dry) taken from the Severn Estuary, but at levels that have made no measurable contribution to gamma whole-body dose rates—the controlling feature of this type of contamination—and at Hinkley, ^{65}Zn has been detected in seaweed taken from the vicinity of the outfall (0.1–0.5 pCi/g wet). Two of the other three stations have only recently become operational. The critical pathway for human exposure at two of the sites is predicted to be external radiation from silt contaminated by activated corrosion products. At the three other sites, the critical pathway is irradiation of the human intestinal tract by neutron activation products concentrated in fish flesh (Preston, 1966).

Humboldt Bay The nuclear power station on Humboldt Bay in northern California began operating in 1962. The quantities of radionuclides discharged to the estuary each year with the cooling water have been substantially below the authorized limit of 20 Ci (0.1 pCi/ml). The nuclides of principal interest in the discharge are ^{65}Zn, ^{60}Co, ^{54}Mn, and ^{137}Cs. A preoperational survey and assessment indi-

TABLE 3 Permissible Discharge Rate of ^{65}Zn from the Bradwell Power Station to the Blackwater Estuary[a]

Preoperational Estimate		Follow-up Assessment	
Estimated concentration of ^{65}Zn in estuary per curie released per day	1.5×10^{-7} μCi/ml	Average ^{65}Zn discharge per month	16.6 mCi
Concentration factor–oyster flesh/seawater	100,000	Average ^{65}Zn concentration in oyster flesh at nearest commercial bed	3.4 pCi/g
Daily intake ^{65}Zn in 75 g of oysters	1.1 μCi	ICRP maximum permissible daily intake	0.22 μCi
ICRP maximum permissible daily intake	0.22 μCi	Maximum daily intake of oyster flesh	75 g/day
Estimated maximum daily discharge rate	0.2 Ci	Maximum permissible concentration of ^{65}Zn in oyster flesh	2,900 pCi/g
		Calculated maximum daily discharge rate	0.5 Ci

[a]Adapted from Preston (1967).

TABLE 4 Comparison of Critical Parameters for Five Civil Nuclear Power Station Sites[a]

Parameters	Central Electricity Generating Board Site				
	Berkeley	Hinkley	Dungeness	Sizewell	Wylfa
Site dilution factor	10	10^2	10^3	10	10^2
Water–μCi/ml/Ci of discharge/day	10^{-7}	10^{-8}	10^{-9}	10^{-7}	10^{-8}
Nuclides	^{65}Zn, ^{60}Co	^{65}Zn, ^{60}Co	^{65}Zn, ^{60}Co	^{65}Zn, ^{60}Co	^{60}Co, ^{65}Zn, ^{32}P
Material	Silt and salmon flesh	Silt and fish flesh	Fish flesh	Fish flesh	Fish and shellfish
Concentration factor	10^4	10^4	10^4	10^4	10^4
Population size	10	10	10	10	10
Critical organ	Total body	Total body	Gastrointestinal tract	Gastrointestinal tract	Bone, gastrointestinal tract
Ingestion rate (g/day)	23	195	807	130	205
Exposure (hr/yr)	340 (silt)	750 (silt)	–	–	–

[a]Adapted from Preston (1966, p. 735) and Mitchell (1967b).

cated that the critical exposure pathway would be via the accumulation of ^{65}Zn in oysters that are grown commercially in Humboldt Bay. Postoperational surveys have never shown levels of radionuclides in oysters of the Bay that were significantly above background. Under experimental conditions, oysters have been held directly in the plant discharge water, and at one time, their concentration of ^{65}Zn reached 175 pCi/g. The level has since declined (Salo, 1968). Inasmuch as the shellfish consumed by the public have only background amounts of the radionuclides, the radiation dose is negligible.

Tarapur A preoperational assessment of critical pathways and radionuclides has been carried out for the Indian power station at Tarapur (Kamath *et al.*, 1966). Neutron activation products (mainly ^{65}Zn) in marine fish consumed by fishermen and families are expected to contribute most to human exposure.

NUCLEAR-POWERED VESSELS

Studies have been made of the amounts and the fate of waste radionuclides discharged to coastal waters by U.S. Navy nuclear-powered submarines and ships at 13 U.S. docking facilities (Vaughan and Miles, 1966) and of conditions in harbors used by the civilian ship NS *Savannah* (Flora and Wukasch, 1966). (Discharges from the U.S. Naval ships are listed in Chapter 2.) No measurable transfer to man of the discharged radionuclides has occurred, and external radiation dose rates near the facilities have shown no measurable increase.

The accidental loss of the *Thresher* in 1964 was followed by extensive radiation monitoring in the vicinity of the hulk. No detectable escape of radionuclides into the sea was found, and no human exposure resulted.

SOLID-WASTE DISPOSAL

United States Sea disposal of contaminated solid wastes was carried out along both the Pacific and Atlantic coasts of the United States during the years 1946–1963. The total quantity disposed in the Pacific was approximately 15,000 Ci, and in the Atlantic, 46,000 Ci (Belter, 1965). Radiation monitoring of the two areas (Pneumo Dynamics Corporation, 1961; Brown *et al.*, 1962) has been carried out. No detectable concentrations of waste radionuclides were found in seawater or marine organisms.

United Kingdom Contaminated solid wastes containing approximately 1,500 Ci of beta-gamma radionuclides are disposed of yearly by the U.K. Atomic Energy Authority into the Atlantic deeps at a depth of not less than 1,500 fathoms. The amounts of waste disposed are considered to be too small to have an appreciable effect on the environment; therefore no radiation monitoring is undertaken (U.K. Atomic Energy Authority, 1966).

Asia Some radioactive wastes have been disposed of in the North Pacific near Japan. One of several measurements made by Akiyama (1965) suggested the presence of artificial contamination near the bottom in the vicinity of the disposal site. The measurement was only slightly above the expected background level, and one would not expect it to have any radiological implications.

CONCLUSIONS

Over the past decade, the use of atomic energy for peaceful purposes, and especially the production of electric power, has moved from the stage of planning and technical development to the stage of demonstration and competitive application. This application has been accompanied by the discharge of low-level radioactive waste into rivers and directly into the sea. In every case, the proposed discharge has received careful study in advance of operations, and prudent restrictions have been specified on the kinds and amounts of radioactive materials that could be released. Follow-up surveys show that the restrictions have been entirely adequate to keep human exposure well within the guidelines specified by the ICRP and FRC. A continuation of the policies and practices concerning the control of low-level waste disposal that have been established during these formative years should assure that radioactive contamination of the marine environment will not reach unacceptable levels.

SUMMARY

The addition of artificial radioactive materials to the marine environment results in some added radiation exposure to people who use the sea and its products. The magnitude of the additional exposure that results depends upon many complex relationships, however, and involves the kinds and quantities of radioactive materials added and the manner and place of their introduction.

Over the past quarter century, the major sources of artificial radioactive materials to the sea have been worldwide fallout from the testing of nuclear devices in the atmosphere and the chronic discharge of low-level wastes from operating reactors and fuel processing plants. Much less significant additions have resulted from nuclear detonations below the surface of the sea, from the disposal of low-level waste in packages, and from the inadvertent loss of radioactive materials.

Radioactive contaminants can follow a variety of pathways in the sea that may bring them into contact with man and contribute to the radiation exposure that he receives from the environment as a whole. Only careful evaluation of each individual situation can determine the most important or critical pathway, but the following ones are likely to be of greatest importance:

The accumulation of certain radionuclides in fish, shellfish, and seaweeds that are eaten by man in substantial quantities

The deposition of radioactive particles on the seabed in places where people may contact them either in the course of their occupations or in pursuit of recreation

The adsorption of certain radionuclides on fishing gear.

A large number of other pathways have been identified or postulated, but at this point in time their contribution to the radiation exposure of people has been much smaller than the ones listed above.

The significance of the radiation exposure contributed by all of the pathways is dependent on the intensity (concen-

tration) of the source, on the frequency and length of time that people are exposed to the source, and on the number of people that may be exposed. It is generally held that any exposure to ionizing radiation entails at least some small risk of a deleterious biological effect. Therefore, the radiation dose must be held to such a low level that the very small risk involved is acceptable to society as a whole in relation to the benefits derived from the use of atomic energy. The Federal Radiation Council, the National Council on Radiation Protection and Measurements, the International Commission on Radiological Protection, and other authoritative groups have recommended doses that they believe should be considered as the maximum acceptable for continuous exposure. In all cases, they recommend restricting exposure to the lowest practicable level compatible with economic and social considerations. For radioactive materials deposited in the body, dose rates are related to a permissible body (or organ) burden for specific radionuclides, and these in turn are related to a permissible continuous daily intake sufficient to establish and maintain that body burden.

When an introduction of radioactive material into the marine environment is being considered, it is now conventional to identify the probable critical pathway of exposure and the individuals or critical population likely to receive the greatest exposure. Preoperational guides are then calculated that establish the maximum allowable rates of introduction of specific radionuclides. The preoperational guides are based on the critical exposure pathways and on some fraction of the internationally recognized dose limits. Following the actual introduction of potentially significant quantities of radioactive materials, a re-evaluation of the probable dose to man can and should be made on the basis of actual measurements of the concentrations of radionuclides in the environment. Follow-up evaluations should be based on measurements of the materials directly responsible for human exposure—the species of fish, shellfish, and seaweed actually consumed by the public, the beaches used by the public, and the gear handled by the fishermen. The follow-up evaluations have almost always shown that the preoperational predictions were highly pessimistic and thus that the tentative discharge guides were much more restrictive than actually necessary to maintain the radiation dose to people within the prescribed limits.

The radiation dose received by people from worldwide fallout via food from the sea is very small both in magnitude and in relation to that received from the terrestrial environment. Small population groups that depend on fish and other marine organisms could have received at most about 0.4 mrem per year to the whole body and 30 mrem per year to parts of the skeleton.

The large nuclear installations of the U.K. Atomic Energy Authority at Windscale on the Irish Sea and of the U.S. Atomic Energy Commission at Richland on the Columbia River discharge far greater quantities of radionuclides into the sea than do nuclear power reactors. The critical exposure pathway for the Windscale plant is the accumulation of ^{106}Ru by seaweed that is eaten in quantity by a small population group in a distant area. The resulting radiation dose amounts to about 40 percent of the recommended limit. The critical exposure pathway for the Richland plant (Hanford) is the concentration of ^{32}P and ^{65}Zn by fish in the Columbia River and oysters grown near the mouth of the river. The marine pathway leads to a dose to people who eat oysters of less than 1 percent of the recommended limit.

Discharges to the sea from power-producing reactors have resulted in doses to the public of much less than 1 percent of the recommended limit. At many sites, the concentrations of reactor-produced nuclides in the marine environment are so low that they can be measured only with sophisticated techniques. The radionuclides from power reactors that appear to be of greatest interest in relation to human exposure are the neutron activation products ^{65}Zn, ^{60}Co, and ^{32}P, because of their accumulation in marine organisms used for food.

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